Measurement of natural ($^{40}\text{K}$, $^{226}\text{Ra}$ and $^{232}\text{Th}$) and anthropogenic ($^{137}\text{Cs}$) gamma radiation and radiological hazard assessment in beach sand samples collected from Ao Phrao at Koh Samet in Rayong province (Thailand)

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Abstract. In the present research, the main objectives were to measure and analyze the specific activities of natural ($^{40}\text{K}$, $^{226}\text{Ra}$ and $^{232}\text{Th}$) radionuclides in 30 beach sand samples collected from Ao Phrao after the Rayong oil spill disaster. This huge Petroleum Authority of Thailand (PTT) oil spill occurred on July 27, 2013, in the Gulf of Thailand nearby Ao Phrao at Koh Samet in Rayong province in eastern region of Thailand. At the same time, anthropogenic ($^{137}\text{Cs}$) radionuclides accumulated in the studied area was also studied and presented. High-purity germanium (HPGe) detector with a massive lead shield, some standard reference material sources and gamma spectrometry technique, were used to measure and analyze all of the experimental results. The frequency distribution of specific activities of $^{40}\text{K}$, $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{137}\text{Cs}$ were also evaluated and found to be the asymmetrical distribution. The median values of $^{40}\text{K}$, $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{137}\text{Cs}$ were 173.55 ± 67.04, 10.76 ± 1.71, 6.35 ± 0.75 and 0.24 ± 0.10 Bq/kg, respectively. Furthermore, the associated radiological hazard indices for the studied area were calculated and compared to nationally and internationally recommended values. It was found that the beach sand samples from the study area provide no excessive exposures created from the oil spill for inhabitants. Moreover, the radioactive contour maps of the investigated area were also imaged and presented in this study. The data presented in the study are importance since they create the baseline of the radiological hazard data and mapping of the investigated area in the future.

1. Introduction
A pipeline owned by the Petroleum Authority of Thailand, Global Chemical Plc. (PTTGC) leaked 50,000 L of crude oil into the Gulf of Thailand off the coast of Koh Samet, a popular tourist island in Mueang Rayong District of the Rayong Province on 27 July 2013. Tourists in the area were evacuated and emergency response teams entered the affected areas. Dispersants, chemicals that break up oil, were applied to the oil spill, causing much of the oil to mix with the seawater and sink to the bottom of the ocean. The remaining oil flowed to Ao Phrao which is located in western side of the island, a
popular tourist beach on Koh Samet, where clean-up crews worked to remove the oil. While the aesthetic condition of the beach was restored by clean up teams within the first two weeks, the health, social, and economic impacts still affect local communities currently six months after the spill. After the oil spill, the tourism and fishing industries suffered due to consumers’ concern over contaminated beaches and seafood. Hence, community members have raised concerns over the amount of information shared by PTTGC about the amount of oil spilled and the procedure in which dispersants were added [1].

Oil spills negatively impact the economy through the destruction of natural resources and livelihoods of the people in contaminated areas. A consequence of an acute effect of oil exposure, then, might be the sudden increasing of the concentration of natural radionuclides which were originated at the beginning the earth and mixed with the crude oil. Hence, there are many groups of researchers and scientists throughout the world are trying to study the effect of oil and gas activities on the radiation level of the environment. For example, the activity concentration of natural radionuclides $^{238}$U, $^{226}$Ra and $^{232}$Th were determined in Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) sludge waste. It was found that sludge waste from oil and gas industry is one of the major sources of $^{226}$Ra in the environment [2]. Enhanced oil and gas production activities have resulted in the increased production of Technologically Enhanced Naturally Occurring Nuclear Materials (TENORMs). This has raised a radiological concern for workers, the public and the environment [3]. A study of the external background radiation in areas affected by Naturally Occurring Radioactive Materials (NORM) generated by oil and gas industrial activities has been performed in the Kingdom of Bahrain. In this framework, two experimental residential areas, Awali and Riffa Views, were selected due to the presence of extensive oil and gas exploration and transportation [4]. There are approximately 40,000 oil fields globally and 6 million people that live or work nearby. Oil extraction can impact local soil, water, and air, which in turn can influence community health [5]. Furthermore, some groups of radioactivity expert were also concern about the consequence effect to our environment, such as environmental impact caused by residues containing high activity concentration of natural radionuclides [6]. The radionuclide activity concentrations in most water samples from three oil mineral leases (OML) 30, 58 and 61 are higher than world average values. Calculated hazard indices and committed effective dose to assess the potential radiological health risk in samples are well above their permissible limit. The sources of water in these oil fields have been polluted and may cause some health hazard to the public users [7]. The contamination is due mainly to unregulated oil and gas production activities leading to oil spills and illegal disposal of contaminated materials, indiscriminate industrial and domestic discharges into water bodies. However, uncontrolled spills and discharges from these activities have left the land desolate, degrading most of its aquifers and surface waters, leaving the indigenes with a Hobson choice, eating and drinking contaminated substances everyday of their lives [8].

In consequence, the research project which concern about the radioactive contaminants after the oil spill at Koh Samet of Rayong province was developed and implemented by group of researchers from Faculty of Science Thaksin University (TSU) and Thailand Institute of Nuclear Technology (Public Organization) (TINT). At the same time the relevant effect and radiological risk were also studied and assessed on some locations in Koh Samet of Rayong province. Then, Ao Phrao, which is located on the western side of Koh Samet and might be affected and contaminated from this oil spill accident was chosen to be our investigated area. In case of the environmental and radiological impacts concern, we would like to measure and evaluate the quantity of specific activities of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides in beach sand samples in this area to make sure that no contamination on the beach. The frequency distribution of specific activities and radiological hazard indices were also studied and calculated for the studied area. Furthermore, the results were used to compare with some research data in Thailand and worldwide evaluations. Moreover, the radioactive contour maps of the investigated area were also created and shown in this study.
2. Experimental

2.1. Sampling and sample preparation
All of 30 beach sand samples collected from Ao Phrao at Koh Samet in Rayong province in eastern region of Thailand as shown in figure 1, dried up at room temperature after collection and sieved through a 2 mm mesh-sized sieve to remove stone, grass and other macro-impurities. The samples were homogenized and then dried in a temperature-controlled furnace at 105 °C for 5 h for removing moisture. Finally, the dried samples were weighed and stored in gastight, radon impermeable, cylindrical polyethylene plastic containers (7.5 cm height and 8.2 cm diameter). The containers were hermetically sealed to prevent the escape of gaseous $^{222}$Rn and $^{220}$Rn from the samples and kept aside for about 1 month to ensure radioactive equilibrium. After that period, the samples were measured using gamma spectrometry technique.

![Figure 1. Map of Koh Samet in Rayong province and sampling locations.](image)

2.2. Measurement and analysis
The specific activities of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides in all of 30 beach sand samples collected from Ao Phrao at Koh Samet in Rayong province were determined by using a high-purity germanium detector (HPGe, EG&G ORTEC Model GEM) and gamma spectrometry analysis system at advanced laboratory, Thailand Institute of Nuclear Technology (Public Organization) (TINT). 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 IAEA Soil-375 reference materials (International Atomic Energy Agency IAEA, Vienna, Austria). Counting time interval was 10800 s. The background spectrum was recorded immediately after or before the sample counting. 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 formulae
The potential radiological hazards for individuals in the investigated area could be evaluated from outdoor air absorbed gamma dose rate (D), radium equivalent activity (Ra<sub>eq</sub>), external hazard index (H<sub>ex</sub>) and annual effective dose rate (AED<sub>out</sub>). All four radiological hazard indices were studied and presented by the authors’ earlier work [9, 10] and given in equation (1), (2), (3) and (4), respectively.

\[ D = 0.461C_{Ra} + 0.623C_{Th} + 0.0414C_K, \]  \hspace{1cm} (1)

\[ Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \]  \hspace{1cm} (2)

\[ H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \]  \hspace{1cm} (3)

\[ AED_{out} (\text{mSv/y}) = D \ (\text{nGy/h}) \times 8760 \ h \times 0.2 \times 0.7 \ (\text{Sv/Gy}) \times 10^{-6} \]  \hspace{1cm} (4)

where C<sub>Ra</sub>, C<sub>Th</sub> and C<sub>K</sub> are the medium values of specific activity of 226Ra, 232Th and 40K in Bq/kg, respectively. All four radiological hazard indices and their average values were calculated and compared with the recommended values reported by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR).

3. Results and Discussions

3.1. Frequency distribution of specific activities of natural (40K, 226Ra and 232Th) and anthropogenic (137Cs) radionuclides
The frequency distribution of specific activities of natural (40K, 226Ra and 232Th) and anthropogenic (137Cs) radionuclides in 30 beach sand samples collected from Ao Phrao beach in Rayong province were studied and analyzed by using a statistic computer program and all results were presented in the following figures 2 - 5. Furthermore, all statistic values which were given from the studying were also shown in table 1.

![Figure 2](image1.png)  \hspace{1cm} Figure 2. Frequency distribution of specific activity of 40K in Ao Phrao beach sand samples.

![Figure 3](image2.png)  \hspace{1cm} Figure 3. Frequency distribution of specific activity of 226Ra in Ao Phrao beach sand samples.
Figure 4. Frequency distribution of specific activity of $^{232}$Th in Ao Phrao beach sand samples.

Figure 5. Frequency distribution of specific activity of $^{137}$Cs in Ao Phrao beach sand samples.

Table 1. Statistic values of frequency distribution of specific activities of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides in 30 beach sand samples collected from Ao Phrao in Rayong province (Thailand).

| Statistic values | $^{40}$K | $^{226}$Ra | $^{232}$Th | $^{137}$Cs |
|------------------|---------|-----------|-----------|-----------|
| Mean Bq/kg       | 194.55  | 11.31     | 6.25      | 0.24      |
| Median Bq/kg     | 173.55  | 10.76     | 6.35      | 0.24      |
| Mode Bq/kg       | 173.55  | 9.04      | 4.30      | 0.22      |
| Skewness         | 0.61    | 0.59      | -0.45     | 0.59      |
| Kurtosis         | -1.02   | 0.37      | -0.67     | -         |
| Minimum value Bq/kg | 110.22  | 5.37      | 4.15      | 0.22      |
| Maximum value Bq/kg | 340.74  | 19.68     | 7.93      | 0.27      |

From figures 2 - 5 and all calculated statistic values in table 1, it was found that the frequency distribution of specific activities of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides in 30 beach sand samples collected from the investigated area, were asymmetrical distribution with the skewness of 0.61, 0.59, -0.45 and 0.59, respectively. For this reason, the median values of $^{40}$K, $^{226}$Ra, $^{232}$Th and $^{137}$Cs which were 173.55 ± 67.04, 10.76 ± 1.71, 6.35 ± 0.75 and 0.24 ± 0.10 Bq/kg, for the studied area, should be selected and used to evaluate the corresponding radiological hazard evaluation in the studied area.

3.2. Median values of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides and comparison

We can see that the specific activities of $^{40}$K in Ao Phrao were mostly lower than almost all of research data in Thailand (except in Pakmeng and Prapat beaches in Phuket and Ranong province, respectively), OAP research data and worldwide mean. Moreover, the specific activities of $^{226}$Ra and $^{232}$Th were mainly lower than all research data in Thailand (but higher than the data of Ao Nang and Napharat Thara beaches in Krabi province), OAP research data and worldwide mean as presented in table 2.

3.3. Radiological hazard evaluation in the investigated area

Moreover, four radiological hazard indices as shown in equations (1) to (4) for the investigated area were calculated and presented in table 3 by the median values of specific activities of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) radionuclides. Moreover, the results were also compared with some research data in Thailand and UNSCEAR as shown in the same table.
Table 2. Comparison of median values of natural ($^{40}$K, $^{226}$Ra and $^{232}$Th) and anthropogenic ($^{137}$Cs) radionuclides in 30 beach sand samples collected from Ao Phrao in Rayong province with some research data in Thailand, Office of Atoms for Peace (OAP) annual report data and worldwide mean.

| Locations | Specified Activity (Bq/kg) | $^{40}$K | $^{226}$Ra | $^{232}$Th | $^{137}$Cs |
|-----------|---------------------------|---------|-----------|-----------|-----------|
| Patong beach [11] (Phuket province) | | 3538.09 ± 959.56 | 29.72 ± 11.48 | 32.71 ± 7.51 | 3.28 ± 1.03 |
| Naiyang beach [11] (Phuket province) | | 1648.27 ± 702.98 | 14.62 ± 6.54 | 18.10 ± 4.22 | 1.19 ± 0.66 |
| Takua Pa and Thai Muang beaches [12] (Phang Nga province) | | 1069.99 ± 119.43 | 40.39 ± 4.30 | 41.15 ± 3.65 | N.A. |
| Ao Nang beach [13] (Krabi province) | | 330.73 | 4.60 | 5.78 | N.A. |
| Noppharat Thara beach [14] (Krabi province) | | 307.60 | 5.31 | 5.28 | N.A. |
| Pakmeng beach [15] (Trang province) | | 57.3 | 23.1 | N.A. |
| Chaweng beach [16] (Surat Thani province) | | 373.30 | 18.85 | 23.53 | N.A. |
| Praphat beach [17] (Ranong province) | | 63.86 ± 2.28 | 39.98 ± 1.02 | 17.46 ± 0.63 | N.A. |
| Ao Phrao (Rayong province)$^a$ | | 173.55 ± 67.04 | 10.76 ± 1.71 | 6.35 ± 0.75 | 0.24 ± 0.10 |
| OAP (Southern region of Thailand)[18] | | 511.04 ± 7.04 | 171.55 ± 3.13 | 211.19 ± 1.98 | 1.13 ± 0.49 |
| Worldwide mean [19] | | 400 | 35 | 30 | N.A. |

$^a$ Present study

Table 3. Comparison between four radiological hazard indices for Ao Phrao in Rayong province with some researches data in Thailand and UNSCEAR.

| Locations | $D$ (nGy/h) | $Ra_{eq}$ (Bq/kg) | $H_{ex}$ | AED$_{out}$ (mSv/y) |
|-----------|------------|------------------|---------|-------------------|
| Patong beach (Phuket province) | 180.56 ± 49.70 | 348.93 ± 96.11 | 0.94 ± 0.26 | 0.22 ± 0.06 |
| Naiyang beach (Phuket province) | 86.25 ± 34.75 | 167.42 ± 66.70 | 0.45 ± 0.18 | 0.11 ± 0.04 |
| Takua Pa and Thai Muang beaches (Phang Nga province) | 88.55 ± 9.20 | 181.62 ± 18.71 | 0.49 ± 0.05 | 0.11 ± 0.01 |
| Ao Nang beach (Krabi province) | 19.41 | 38.33 | 0.10 | 0.02 |
| Noppharat Thara beach (Krabi province) | 18.47 | 36.55 | 0.10 | 0.02 |
| Pakmeng beach (Trang province) | 17.32 | 37.38 | 0.10 | 0.02 |
| Chaweng beach (Surat Thani province) | 38.80 | 81.24 | 0.22 | 0.05 |
| Praphat beach (Ranong province) | 31.95 ± 0.96 | 69.86 ± 2.10 | 0.19 ± 0.01 | 0.04 ± 0.01 |
| Ao Phrao (Rayong province)$^a$ | 16.10 ± 4.03 | 33.20 ± 7.94 | 0.09 ± 0.02 | 0.02 ± 0.01 |
| UNSCEAR [19-21] | 55 | 370 | 1 | 0.48 |

$^a$ Present study
According to four radiological hazard indices in the studied area as shown in table 3, we can see that the values of D was 16.10 ± 4.03 nGy/h which was lower than all research data in Thailand and lower than 55 nGy/h as reported by UNSCEAR. The values of Ra_{eq} in Ao Phrao in Rayong province was lower than all research data in Thailand and in the same level of Ao Nang, Noppharat Thara and Pakmeng beaches in Krabi and Trang provinces which is less than 370 Bq/kg, which are the acceptable value for safe use. The values of H_{ex} obtained in this study was equal to 0.09 ± 0.02 which were less than unity and also lower than in all research data in Thailand and UNSCEAR data. The calculated AED_{eq} for the Ao Phrao was also determined and equal to 0.02 ± 0.01 mSv/y which lower than the worldwide average value of 0.48 mSv/y as presented by UNSCEAR and the maximum dose constraint value of 1.0 mSv/y as presented by International Commission on Radiological Protection (ICRP) [22].

3.4. The radioactive contour maps (RCM) of the investigated area
Consequently, radioactive contour maps (RCM) of natural (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic (^{137}Cs) radionuclides in 30 beach sand samples collected from Ao Phrao in Rayong province were created by using computer program and shown in the following figures 6 - 9, respectively.

**Figure 6.** Radioactive Contour Maps (RCM) of ^{40}K in Ao Phrao beach sand samples.

**Figure 7.** Radioactive Contour Maps (RCM) of ^{226}Ra in Ao Phrao beach sand samples.

**Figure 8.** Radioactive Contour Maps (RCM) of ^{232}Th in Ao Phrao beach sand samples.

**Figure 9.** Radioactive Contour Maps (RCM) of ^{137}Cs in Ao Phrao beach sand samples.

We can see that the radioactive contour maps (RCM) of natural (^{40}K, ^{226}Ra and ^{232}Th) and anthropogenic (^{137}Cs) radionuclides for the investigated area as shown in figures 6 - 9, were created and appeared in normal level.
4. Conclusions
The median values of specific activity of natural (\(^{40}\)K, \(^{226}\)Ra and \(^{232}\)Th) and anthropogenic (\(^{137}\)Cs) radionuclides in 30 beach sand samples collected from Ao Phrao in Rayong province were comparable to research data in Thailand, OAP research data and worldwide mean. Hence, all four values of radiological hazard indices in the studied area which were \(D, R_{eq}, H_{\alpha}\) and \(AED_{out}\) were lower than the recommend values which reported by UNSCEAR. All of radioactive contour maps (RCM) were in the same level of the background radiation. For this reason, the radioactive contaminants after the oil spill in the Gulf of Thailand on July 27, 2013 would not have a significant effect on beach sand samples of the investigated area. From a useful perspective, the values of natural and anthropogenic radionuclides found can be used as a database useful to compare, monitor and quantify possible changes in ecological radioactivity due to nuclear, modern and other human actions.

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