

GAMMA RADIATION HAZARD EVALUATION AND EXCESS LIFETIME CANCER RISK ASSESSMENT IN SURFACE SOIL SAMPLES IN PLUAK DAENG DISTRICT IN RAYONG PROVINCE, THAILAND

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Abstract

The specific activity of natural (^{226}Ra , ^{232}Th , and ^{40}K) and anthropogenic (^{137}Cs) radionuclides in 20 surface soil samples collected from Pluak Daeng district in Rayong province (Thailand) were measured and analysed systematically by using High Purity Germanium (HPGe) detector and gamma-ray spectrometry system. By using the statistic computer program, the specific activity distribution of required radionuclides were studied and presented in this study. It was found that all of distributions of ^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs were asymmetrical with the median values of 75.14 ± 1.74 , 29.61 ± 0.83 , 186.51 ± 7.09 , and 1.20 ± 0.22 Bq/kg, respectively. Hence, the median values of ^{226}Ra , ^{232}Th , and ^{40}K would be chosen and used to evaluate the consequential gamma dose and the corresponding radiation hazard for the investigated area and were found to be within the permissible limits. Furthermore, the excess lifetime cancer risk (ELCR(outdoor)) would also be evaluated for the studied area. The results were also compared with the Office of Atoms for Peace (OAP) annual report data, Thailand and global radioactivity measurement and calculations. Moreover, the radioactive contour maps (RCM) of the investigated area were also created and presented in this paper.

Keywords: Specific activity, surface soil, gamma radiation hazard, excess lifetime cancer risk, radioactive contour map

Introduction

Naturally occurring radioactive materials (NORMs) are inherent in many geologic materials and consequently encountered during geologically related activities. NORMs encountered in oil and gas exploration, development and production operations originate in subsurface formations that may contain radioactive materials, such as uranium and thorium

and their daughter products, ^{226}Ra and ^{228}Ra . This can be brought to the surface in the water that is produced in conjunction with oil and gas (Ajayi *et al.*, 2009). Wastes associated with various industrial activities were enhanced levels of natural radioactivity which called Technological Enhanced Naturally Occurring Radioactive Materials (TENORMs). Enhanced oil

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and gas production activities have resulted in the increased production of TENORMs. This has raised a radiological concern for workers, the public and the environment (Nabhani *et al.*, 2017). Recently, attention was focused on the environmental and health impacts from the uncontrolled release of TENORMs wastes. From the natural risk point of view, it is necessary to know the dose limits of public exposure and to measure the natural environmental radiation level provided by TENORMs to estimate human exposure to natural radiation sources (Nabhani *et al.*, 2016; Husain and Sakhnini, 2017; Jodlowski *et al.*, 2017). Rayong province is located about 220 km from Bangkok and also located relatively close to Thailand's offshore gas fields in the Gulf of Thailand and to major gas consuming centres such as Bangkok. This led to Thailand's three major offshore gas pipelines being constructed through the region. Partly as a result, the province, and particularly the Map Ta Phut industrial complex, has grown into one of the world's biggest petrochemicals hubs. Over the past 20 years, Rayong province has experienced significant industrial development. We can see that Rayong province is the province with the highest per capita income in Thailand. Unfortunately, this area have no systematic data and information about the baseline level of NORMs and TENORMs (^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs) which are accumulated on its environment such as surface soil, beach sand and water. For this reason, Rayong province should be the interesting investigated area to explore the level of all those radionuclides. This study will focus on Pluak Daeng district which is located in the north western part of Rayong province because of its own size, location and have not many industrial estates in this area. The main objectives of this research is to determine the background level of specific activity in surface soil samples and some radiation indices in Pluak Daeng district. This could be also use to evaluate the risks of radiations from TENORMs wastes that produced from oil and gas industry in Rayong province. This would protect workers in oil and natural gas production industry as well as environment in the investigated area by reducing its activity concentration consequently and the health hazard potential.

Study Area

Pluak Daeng district which is one of eight districts and located in the north western region of Rayong province in Thailand. Neighboring districts are Wang Chan, Ban Khai, Nikhom Phatthana of Rayong Province, Bang Lamung, Si Racha and Nong Yai of Chonburi province. Pluak Daeng district is divided into six sub-districts, which are

further subdivided into 34 administrative villages. The population of Pluak Daeng district is about 70,748 in 2019 (Rayong Provincial Public Health Office, 2019).

Experimental Detail

Entirely 20 surface soil samples were collected (from the ground surface down to the depth of 5-10 cm) from six sub-districts in Pluak Daeng district by random selection. After collection, each surface soil sample was kept in a zip-lock plastic bag and labeled with the name of its location. The Global Positioning System (GPS) tool was used to record the geographical coordinates of the sampling locations. Then, all of surface soil samples were taken to the following process; (i) grinded and dried at room temperature for several days (ii) oven dried a temperature 105°C for 3 h (iii) sieved pass through a 2 mm mesh-sized (iv) packed in a PVC cylindrical container (8.2×7.5 cm) which were externally sealed with a cellophane tape, and (v) stored for approximately one month to ensure the radiation equilibrium of ^{222}Rn and its parent ^{226}Ra in uranium series.

Measurement, Analysis and Study of Frequency Distribution of Specific Activities

The specific activities of natural (^{226}Ra , ^{232}Th , and ^{40}K) and anthropogenic (^{137}Cs) radionuclides in 20 surface soil samples were determined by using a high-purity germanium detector and gamma spectrometry analysis system at advanced laboratory, Thailand Institute of Nuclear Technology (Public Organization) (TINT). Geometric efficiency for soil matrices in the container was determined by the IAEA Soil-375 reference materials (International Atomic Energy Agency IAEA, Vienna, Austria). The specific activity of natural and anthropogenic radionuclides were analyzed and determined by using 609.3 keV photopeak of ^{214}Bi for ^{226}Ra and 583.2 keV photopeak of ^{208}Tl for ^{232}Th . The specific activity of ^{40}K and ^{137}Cs were evaluated from 1,460.8 and 661.7 keV photopeaks of themselves, respectively.

Evaluation of Radiological Hazard Indices and the Excess Lifetime Cancer Risk

In order to estimate the radiological risks, gamma absorbed dose rate (D) in outdoor air (Singh *et al.*, 2005), radium equivalent activity (Ra_{eq}) (UNSCEAR,

2000; Veiga *et al.*, 2006), external hazard index (H_{ex}) (Beretka and Mathew, 1985), annual effective dose rate (AED_{out}) (Singh *et al.*, 2005) and the excess lifetime cancer risk (ELCR(outdoor)) (Qureshi *et al.*, 2014). were calculated as follows :

$$D(nGy\ h^{-1}) = 0.462 C_{Ra} + 0.6044 C_{Th} + 0.0417 C_K \quad (1)$$

$$Ra_{eq} (Bq\ kg^{-1}) = C_{Ra} + 1.43 C_{Th} + 0.077 C_K \quad (2)$$

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

$$AED_{out} = D(nGy\ h^{-1}) \times 8,760\ h \times 0.2 \times 0.7\ Sv\ Gy^{-1} \times 10^{-6} \quad (4)$$

$$ELRC(outdoor) = AED_{out} \times LF \times RF \quad (5)$$

where C_{Ra} , C_{Th} , and C_K are the selected medium values of specific activity of ^{226}Ra , ^{232}Th , and ^{40}K in Bq/kg, respectively. Moreover, the LF and RF are life expectancy of Thai people (75 years) and risk factor ($0.05\ Sv^{-1}$), respectively. All four radiological hazard indices and the ELCR(outdoor) values with their average values were calculated and compared with the recommended values reported by United Nations Scientific Committee on the Effects of

Atomic Radiation (UNSCEAR) (UNSCEAR, 1988; 1993; 2000).

Construction of Radioactive Contour Maps (RCM)

The ArcGIS computer program were employed to create and present all of the radioactive contour maps (RCM) of specific activities of ^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs radionuclides in 20 surface soil samples collected from the investigated area.

Results and Discussions

Frequency Distribution of Specific Activities and Statistic Values

From Figure 1 and all calculated statistic values by using the SPSS computer program, it was found that the frequency distribution of specific activities of ^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs in 20 surface soil samples collected from the investigated area, were asymmetrical distribution with the skewness of 0.91, 2.12, 1.27, and 0.68, respectively. For this reason, the median values of ^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs

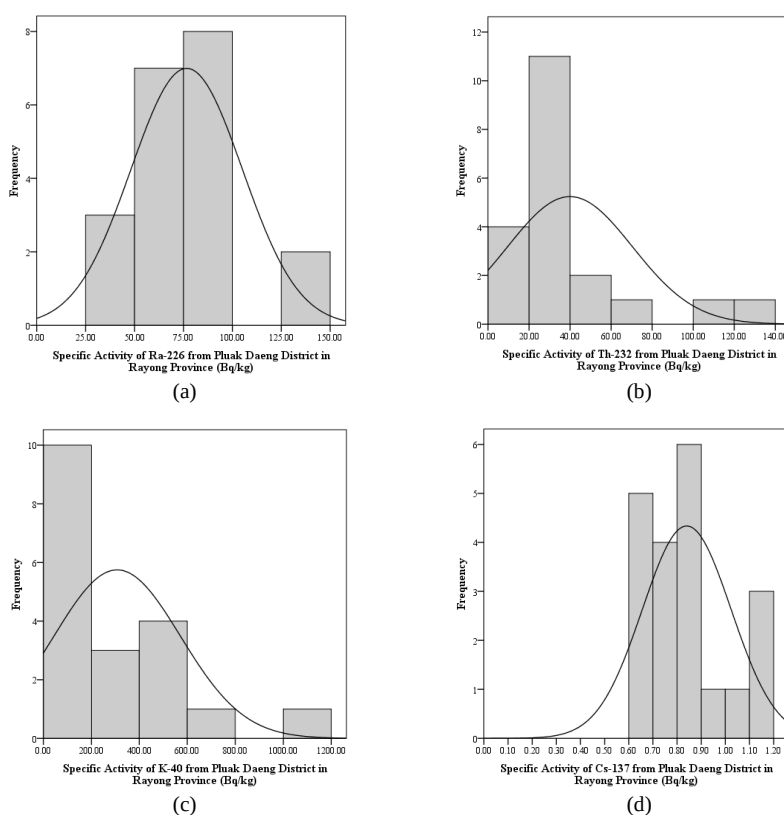


Figure 1. Frequency distribution of specific activities of (a) ^{226}Ra ; (b) ^{232}Th ; (c) ^{40}K and (d) ^{137}Cs in 20 surface soil samples collected from Plauk Daeng district in Rayong province

which were 75.14 ± 1.74 , 29.61 ± 0.83 , 186.51 ± 7.09 , and 1.20 ± 0.22 Bq/kg, should be chosen for calculation the corresponding radiological hazard evaluation in the studied area.

Comparison of the Specific Activities of Natural and Anthropogenic Radionuclides to Thailand Studies and Worldwide Means

The median values of specific activities of ^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs in 20 surface soil samples in the investigated area were also compared with the Office of Atoms for Peace (OAP) annual report data, Thailand and global radioactivity measurement and evaluations as shown in Table 1.

From the result in Table 1, the value of specific activities of ^{226}Ra from the investigated area was higher than the worldwide mean value, Mueang district in Rayong and Krabi provinces. The value of specific activities of ^{232}Th , ^{40}K , and ^{137}Cs from the studied area were mostly lower than the reference values. Furthermore, we can notice that the specific activities value of ^{226}Ra which might be directly caused from oil and gas industry (Hilal *et al.*, 2014) in Rayong province, was close to the value in Surat

Thani province, Krabi province, Nikom Phatthana and Ban Chang districts of Rayong province and about 2.15 times higher than the worldwide mean value.

Radiological Hazard Indices and Excess Lifetime Cancer Risk Evaluation and Comparison

Four radiological hazard indices which are D, Ra_{eq} , H_{ex} , AED_{out} , and ELCR (outdoor) were evaluated for the investigated area by using Equations (1) to (5). Then, all of those calculated values were used to compare to Thailand studies and evaluations, OAP data and UNSCEAR data and presented in Table 2.

From Table 2, we can see that the average values of three from four radiological hazard indices and ELCR(outdoor) in the investigated area were lower than the OAP data and UNSCEAR recommended values. The D in the present study was lower than the OAP data but higher than the UNSCEAR recommended values (about 1.11 times). In addition, the AED_{out} was evaluated and found lower than the UNSCEAR recommended values (about 6.86 times).

Table 1. Comparison of specific activities of ^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs in 6 provinces in the south of Thailand, 3 districts in Rayong province, OAP and worldwide mean values with the present study

Locations	Specific Activity (Bq/kg)			
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Songkhla (Ratanasumneiang <i>et al.</i> , 2010)	107.28 ± 6.74	51.53 ± 5.02	3562.14 ± 223.56	3.05 ± 1.58
Nakhon Si Thammarat (Thongna, 2009).	108.55 ± 31.19	73.95 ± 4.15	4313.18 ± 148.16	4.84 ± 2.59
Surat Thani (Kessaratikoon <i>et al.</i> , 2013).	75.72 ± 5.75	47.77 ± 4.79	2119.10 ± 171.72	3.85 ± 1.67
Phuket (Kessaratikoon <i>et al.</i> , 2015).	240.11 ± 10.79	210.68 ± 9.89	4896.45 ± 288.74	5.91 ± 2.94
Phang-Nga (Kessaratikoon <i>et al.</i> , 2015).	165.44 ± 8.44	160.11 ± 7.92	2879.87 ± 225.50	5.76 ± 2.49
Krabi (Kessaratikoon <i>et al.</i> , 2015).	65.75 ± 4.95	40.69 ± 3.96	802.58 ± 115.25	2.96 ± 1.59
Mueang District (Rayong) (Kessaratikoon <i>et al.</i> , 2018)	49.58 ± 1.38	16.67 ± 0.65	297.20 ± 8.50	1.41 ± 0.40
Nikom Phatthana and Ban Chang Districts (Rayong) (Kessaratikoon <i>et al.</i> , 2019)	96.65 ± 2.16	36.73 ± 1.06	423.75 ± 11.30	1.32 ± 0.24
Plauk Daeng District (Rayong)^a	75.14 ± 1.74	29.61 ± 0.83	186.51 ± 7.09	1.20 ± 0.22
OAP (OAP, 1994-2002)	171.55 ± 3.13	211.19 ± 1.98	511.04 ± 7.04	1.13 ± 0.49
Worldwide mean (UNSCEAR, 2000)	35	30	400	N.A.

^a Present study

Table 2. Comparison of Thailand studies and evaluations, OAP data and UNSCEAR recommended values to D, Ra_{eq} , H_{ex} , AED_{out} , and ELCR(outdoor) to the present study

Locations	D (nGy/h)	Ra_{eq} (Bq/kg)	H_{ex}	AED_{out} (mSv/y)	ELCR ($\times 10^{-3}$)
Songkhla	229.03 ± 15.49	455.25 ± 31.13	1.23 ± 0.08	0.28 ± 0.02	1.05 ± 0.07
Nakhon Si Thammarat	274.68 ± 23.10	546.41 ± 48.53	1.48 ± 0.13	0.34 ± 0.03	1.26 ± 0.11
Surat Thani	152.90 ± 12.81	308.44 ± 25.97	0.83 ± 0.07	0.19 ± 0.02	0.70 ± 0.06
Phuket	446.92 ± 23.19	922.99 ± 47.37	2.49 ± 0.13	0.55 ± 0.03	2.06 ± 0.11
Phang-Nga	295.24 ± 18.16	616.14 ± 37.13	1.66 ± 0.10	0.36 ± 0.02	1.36 ± 0.08
Krabi	89.16 ± 9.53	186.35 ± 19.50	0.50 ± 0.05	0.11 ± 0.01	0.41 ± 0.04
Mueang District (Rayong)	45.54 ± 1.39	96.30 ± 2.95	0.26 ± 0.01	0.06 ± 0.01	0.21 ± 0.01
Nikom Phatthana and Ban Chang Districts (Rayong)	84.98 ± 2.12	181.80 ± 4.55	0.49 ± 0.01	0.10 ± 0.01	0.39 ± 0.01
Plauk Daeng District^a (Rayong)	60.81 ± 1.61	131.84 ± 3.46	0.36 ± 0.01	0.07 ± 0.01	0.28 ± 0.01
OAP Data	231.81 ± 2.97	512.90 ± 6.50	1.39 ± 0.02	0.28 ± 0.01	1.07 ± 0.01
UNSCEAR (UNSCEAR, 1988; 1993; 2000)	55	370	1	0.48	1.80

^a Present study

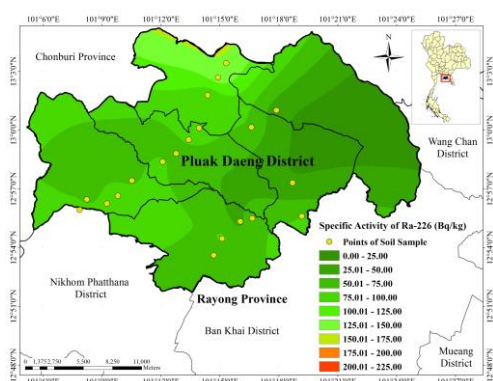


Figure 2. Radioactive Contour Maps (RCM) of ^{226}Ra in surface soil samples of Pluak Daeng district in Rayong Province

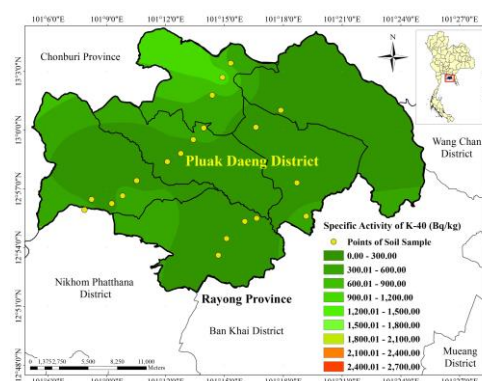


Figure 4. Radioactive Contour Maps (RCM) of ^{40}K in surface soil samples of Pluak Daeng district in Rayong Province

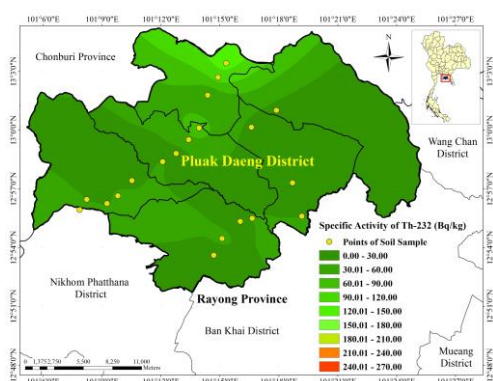


Figure 3. Radioactive Contour Maps (RCM) of ^{232}Th in surface soil samples of Pluak Daeng district in Rayong Province

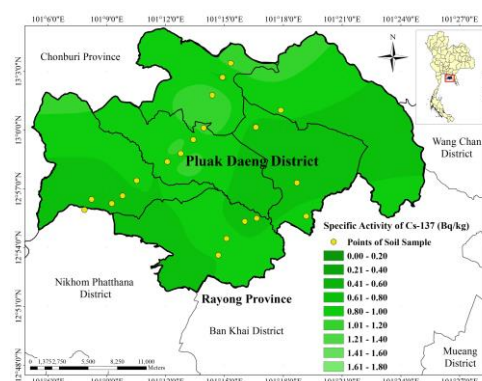


Figure 5. Radioactive Contour Maps (RCM) of ^{137}Cs in surface soil samples of Pluak Daeng district in Rayong Province

The Radioactive Contour Maps (RCM) of the Investigated Area

We can see that the RCM of all 4 radionuclides for the investigated area were created and appeared in green or light green color (as shown in Figures 2-5) which were in normal level and not different to the background radiation.

Conclusions

The median values of specific activity of ^{226}Ra , ^{232}Th , ^{40}K , and ^{137}Cs radionuclides in 20 surface soil samples collected from Pluak Daeng district in Rayong province were in the same range of some research data in Thailand, OAP research data and worldwide mean. Consequently, three from four values of radiological hazard indices (R_{eq} , H_{ex} and AED_{out}) in the studied area were lower than the recommend values which reported by OAP and UNSCEAR. Furthermore, the ELCR (outdoor) was also lower than the reference values of OAP and

UNSCEAR. Moreover, all of RCM were also in the same level of the background radiation. For this reason, the radioactive contaminants from oil and gas extraction industry and human activities would not have a significant effect on surface soil samples in the investigated area. We may use all of values of natural and anthropogenic radionuclides in this study to be the reference or background level for Rayong province. According to the higher value of D in the investigated area than the UNSCEAR recommended value, this area requires more concern, attention and monitoring about the radioactivity level due to modern nuclear technology, Eastern Economic Corridor (EEC) Project and other human actions in the future.

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References

- Ajayi, T.R., Torto, N., Tchokossa, P., and Akinlua, A. (2009). Natural radioactivity and trace metals in crude oils: implication for health. *Environ. Geochem. Health.*, 31(1): 61-69.
- Beretka, J. and Mathew, P.J. (1985). Natural radioactivity of Australian building materials, industrial wastes and by-products. *Health Phys.*, 48(1):87-95.
- Hilal, M.A., Attallah, M.F., Mohamed, G.Y., and Fayed-Hassan, M. (2014). Evaluation of radiation hazard potential of TENORM waste from oil and natural gas production. *J. Environ. Radioact.*, 136:121-126.
- Husain, H. and Sakhnini, L. (2017). Radiological impact of NORM generated by oil and gas industries in the kingdom of Bahrain. *J. Environ. Radioact.*, 167:127-133.
- Jodlowski, P., Macuda, J., Nowak, J., and Dinh, C.N. (2017). Radioactivity in wastes generated from shale gas exploration and production-north-eastern Poland. *J. Environ. Radioact.*, 175-176:34-38.
- Kessaratikoon, P., Boonkrongcheep, R., Benjakul, S., and Youngchauly, U. (2013). Assessment and radioactive contour maps of specific activities of natural (^{40}K , ^{226}Ra and ^{232}Th) and anthropogenic (^{137}Cs) radionuclides in surface soil samples from Surat Thani province, Thailand, [abstract]. The 8th Annual Conference of the Thai Physics Society (SPC 2013): Thai Physics Society on the Road to ASEAN Community; 21-23 March 2013; Chiang Mai, Thailand, p. 340.
- Kessaratikoon, P., Boonkrongcheep, R., Benjakul, S., and Youngchauly, U. (2015). Measurement of specific activity of natural and anthropogenic radionuclides and radiological hazard assessment in surface soil samples collected from three provinces (Phuket, Phang Nga and Krabi) along the Andaman Sea Coast in Southern Region of Thailand. *J. Phys.: Conf. Series.*, 611:1-8.
- Kessaratikoon, P., Boonkrongcheep, R., Eiadkaew, K., and Pholthum, S. (2018). Radiological hazard assessment and excess lifetime cancer risk evaluation in surface Soil Samples Collected from Mueang District in Rayong province, Thailand. In: *Proceeding of the 3rd Environment and Natural Resources International Conference (ENRIC 2018): Global Development with Environmental Sustainability*; November 22-23, 2018; Chonburi, Thailand, p. 72-80.
- Kessaratikoon, P., Jewawongsakul, J., Boonkrongcheep, R., and Pholthum, S. (2019). Radiological hazard assessment and excess lifetime cancer risk evaluation in surface soil samples collected from Ban Chang and Nikhom Phatthana districts in Rayong province, Thailand. *J. Phys. Conf. Series.*, 1380(2019):012104, doi: 10.1088/1742-6596/1380/1/012104.
- Nabhani, K.A., Khan, F., and Yang, M. (2016). Technologically enhanced naturally occurring radioactive materials in oil and gas production: a silent killer. *Process Saf. Environ. Prot.*, 99:237-247.
- Nabhani, K.A., Khan, F., and Yang, M. (2017). Management of TENORMs produced during oil and gas operation. *J. Loss Prev. Process Ind.* 47:161-168.
- Office of Atoms for Peace, [OAP]. (1994-2002). OAP 1994-2002 annual reports. OAP Publication, Bangkok. Available from: Ministry of Science and Technology, Bangkok, Thailand.
- Qureshi, A.A., Tariq, S., Din, K.U., Manzoor, S., Calligaris, C. and Waheed, A. (2014). Evaluation of excessive lifetime cancer risk due to natural radioactivity in the rivers sediments of Northern Pakistan. *J. Radia. Res. Appl. Sci.*, 7:438-447.
- Ratanasumneiang, C., Udomsomporn, S., Benjakul, S. and Kessaratikoon, P. (2010). Measurement of specific activity of natural (^{238}U , ^{232}Th , ^{226}Ra and ^{40}K) and anthropogenic (^{137}Cs) radionuclides in soil from Songkhla province (Thailand). In: *Proceeding of the 20th Thaksin University annual conference: Thai society development with creative research*; 16 -18 September 2010; Songkhla, Thailand, p. 49-61.
- Rayong Provincial Public Health Office. (2019). Population in Pluak Daeng District on Mid-year 2019 (July 1, 2019). Available from: <http://rayonghealth.com/web/stat.php>. Accessed date: November 3, 2019. (in Thai).
- Singh, S., Rani, A., and Mahajan, R.K. (2005). ^{226}Ra , ^{232}Th and ^{40}K analysis in soil samples from some areas of Punjab and Himachal Pradesh, India using gamma ray spectrometry, *Radia. Meas.*, 39(4):431-39.
- Thongna, T. (2009). Natural radioactivity assessment in Nakhon Si Thammarat province, [M.Sc. thesis]. Department of Physics, Faculty of Science, Thaksin University, Songkhla, Thailand, 162p.
- United Nations Scientific Committee on the Effects of Atomic Radiation, [UNSCEAR]. (1988). Sources and effects of ionizing radiation. Report to general assembly, with scientific annexes. United Nations Publications, New York, ISBN 92-1-142143-8, 46p. Available from: United Nations, New York, USA.
- United Nations Scientific Committee on the Effects of Atomic Radiation, [UNSCEAR]. (1993). Sources and effects of ionizing radiation. Report to general assembly, with scientific annexes. United Nations Publications, New York, ISBN 92-1-142200-0, 30p. Available from: United Nations, New York, USA.
- United Nations Scientific Committee on the Effects of Atomic Radiation, [UNSCEAR]. (2000). Sources and effects of ionizing radiation. Report to general assembly, with scientific annexes. United Nations Publications, New York, ISBN 92-1-142238-8, 17p. Available from: United Nations, New York, USA.
- Veiga, R.S.L., Sanches, N., Anjos, R.M.D., Macario, K., Bastos, J., Iguatemy, M., Aguiar, J.G., Santos, A.M.A., Mosquera, B., Carvalho, C., Filho, M.B. and Umisedo, N.K. (2006). Measurement natural radioactivity in Brazilian beach sands. *Radia. Meas.*, 41(2):189-196.