Assessment of radionuclides and heavy metals in marine sediments along the Upper Gulf of Thailand

S Khuntong\textsuperscript{1,5}, C Phaophang\textsuperscript{2} and W Sudprasert\textsuperscript{3}

\textsuperscript{1} Faculty of Science at Si Racha, Kasetsart University Si Racha campus, 199 Sukhumvit Rd., Si Racha, Chonburi 20230 Thailand
\textsuperscript{2} Radioisotope Center, Thailand Institute of Nuclear Technology, Vibhavadi Rangsit Rd., Chatuchak, Bangkok, 10900 Thailand
\textsuperscript{3} Department of Applied Radiation and Isotopes, Faculty of Science, Kasetsart University, Ngam Wong Wan Rd., Chatuchak, Bangkok 10900 Thailand

E-mail: srcstk@ku.ac.th

Abstract. Due to the Fukushima Daiichi nuclear disaster in 2011 and the development of nuclear power plant in neighboring countries such as Vietnam in the near future, radionuclide assessment in marine sediment during 2010 - 2011 may be useful as background levels for radiation protection in Thailand. Marine sediments (10 samples) were collected approximately 1 km away from the coastline along Chonburi to Pattaya, Chonburi Province. The sediments were ground and sieved through 2-mm test sieve after air drying. Radionuclides were measured with a gamma spectrometer equipped with a well-calibrated HPGe detector. The samples were prepared in the same geometry as the reference material. The optimal counting time was 60,000 – 80,000 s for statistical evaluation and uncertainties. No contamination of $^{137}$Cs as an artificial radionuclide was found. Naturally-occurring radionuclides including $^{238}$U, $^{232}$Th and $^{40}$K were found. The mean specific activities of $^{238}$U, $^{232}$Th and $^{40}$K were $44 \pm 10$, $59 \pm 17$ and $463 \pm 94$ Bq/kg in the rainy season (2010); $41 \pm 6$, $50 \pm 9$ and $484 \pm 83$ Bq/kg in the winter (2010), and $39 \pm 6$, $41 \pm 7$ and $472 \pm 81$ Bq/kg in the summer (2011), respectively. The mean specific activities were higher than the values in the UNSCEAR report of 35, 30 and 400 Bq/kg for $^{238}$U, $^{232}$Th and $^{40}$K, respectively. From the measured specific activities, the absorbed dose rate, radium equivalent activity, external hazard index and annual external effective dose rate were calculated in order to assess the health risk. No radiation hazards related to the radioactivity in the sediment were expected. The accumulation of radionuclides varied with the particle size and the organic matter content in the sediment. The accumulation of heavy metals showed similar results to that of the radionuclides in the sediment.

1. Introduction
Radionuclides such as U, Th, Ra, and Am \cite{1} constitute a category of heavy metals in marine environment as they become and cause serious risks to human health and ecosystem \cite{2}. They are generated from two different sources: 1) natural sources which yield a constant activity level and 2) artificial sources which are caused by nuclear weapon tests, nuclear accidents, nuclear power plants, nuclear waste and so on causing a variation in the radioactivity level. Increasing utilization of radioactive

---
\textsuperscript{6} To whom any correspondence should be addressed.
materials for industrial and medical purposes may cause contamination of radionuclides in the marine environment. Daughter decay products (Rn, Ra, Pu and Pb) from natural radioactive series Th-232, U-235 and U-238 and the others (K-40 and Ru-87) are mobilized and accumulated in the marine sediments. Components of the marine environment such as the appearance of the sediment, the organic content, the grain size and geochemical properties (properties of seawater), the benthos and the marine organisms could be affected by the accumulation of radionuclides in marine sediments.

Because of their high molecular weight, the chemical properties of several radionuclides may be similar to heavy metals. They are normally bound to particulate matters and finally settled down and incorporated into the marine sediments when they are released into the aquatic system [3]. Marine sediments are then considered as the secondary source originated from the sinking of the pollutants. Resuspension, bioturbation and diagenesis of sediments naturally can attenuate the pollutants in the sediments [4], [5]. Similar to heavy metals, radionuclides can accumulate in sediments over a long time [6]. The physical, chemical and biological properties of interstitial water and sediments bring about various forms of metals. The mechanisms of metal accumulation in the sediments lead to five geometric forms: (1) exchangeable, (2) bound to the carbonate phase, (3) bound to iron-manganese-oxide, (4) bound to the organic matters and (5) residual metal phase [7].

The study area in this report was located along the Chonburi coastline of Thailand from Ban Suan to Bang Lamung spreading over a 50 km distance. This area constituted mostly the industrialized area, the shipping area and the tourism area. Radioactivity levels from the natural radionuclides and the fission products before Fukushima Daiichi disaster especially I-131, Cs-134 and Cs-137 were evaluated. The data may be used as a guideline for background levels of radionuclides in the marine sediments.

2. Material and method

2.1. Sampling locations and sample preparation
Marine sediments were collected along Chonburi coastline from Ban Suan to Bang Lamung district (part of the upper Gulf of Thailand, a semi-enclosed gulf) over about 50 km distance. Each sampling point was located approximately 1 km away from the shore in three seasons (rainy: September, 2010, winter: December, 2010, and summer: March, 2011). Coordinates were determined by the global positioning system (figure 1). The physical parameters consisting of temperature, electrical conductivity, salinity and dissolved oxygen were measured at each half depth below the sea level by using YSI-60 multimeter. The sediment samples were collected by Ekman dredge grab and some samples were taken by scuba diving. The depth of the sediment cores were between 0 – 10 cm.

After air drying, the dried sediment was ground and sieved through a 2 mm test sieve. The ground sediment was then filled into a plastic cylindrical container with the same geometry as IAEA Soil 6 reference material (7.5 x 7 cm²).

2.2. Classification of sediment texture and organic content
The texture of the sediments and their organic content were characterized since they might affect the deposition of radionuclides and heavy metals in the sediments. Dried sediment (5 g) was heated at 350-440 °C for 24 h to remove organic matter. The percentage of organic content was determined by the weight difference [8].

2.3. Benthos sampling and identification
Fractions of the sediment samples were washed and collected for benthos by sequential sieving through 2 mm, 850 µm and 425 µm sieves. The benthos were removed by forceps and immediately placed under a stereomicroscope for taxonomic identification. If the benthos could not be immediately identified, they were stored in 10% formalin until identification.
2.4 Measurement of radioactivity and determination of health risks

Radionuclides in the sediments were measured with a gamma spectrometer equipped with HPGe detector (Ortec GWL-120230 MJ-GWL). The energy and efficiency of the detector were calibrated with IAEA Soil 6 reference material [9].

For statistical confidence, the counting time of marine sediment was optimized to 60,000-80,000 s. The radioactivity was calculated by:

\[
A = \frac{R_n}{\varepsilon P_\gamma W}
\]  

where \(A\) is the activity of the radionuclides (Bq/kg); \(R_n\) is the counting rate (cps); \(\varepsilon\) is the detector efficiency; \(P_\gamma\) is the absolute probable decay of gamma energy \(\leq 1\), and \(W\) is the sediment weight (g).

To assess the radiation hazards associated with the radioactivity in the sediments, the absorbed dose rate in air \(D\), the radium equivalent activity \(R_{\text{eq}}\), the external hazard index \(Hex\), and the annual external effective dose rate \(AED_\text{out}\) were calculated from activities of U-238, Th-232 and K-40 using equations 2 - 5 [10].

\[
D \text{ (nGy/h)} = 0.0417C_K + 0.462C_U + 0.604C_{Th} \tag{2}
\]

\[
R_{\text{eq}} \text{ (Bq/kg)} = 0.077C_K + C_U + 1.43C_{Th} \tag{3}
\]

\[
Hex = \frac{C_K}{4810} + \frac{C_U}{370} + \frac{C_{Th}}{259} \tag{4}
\]

\[
AED_\text{out} \text{ (mSv/y)} = D \times 8760 \times 0.2 \times 0.7 (\text{Sv/Gy}) \times 10^{-6} \tag{5}
\]

where \(C_K\), \(C_U\) and \(C_{Th}\) are specific activities (Bq/kg) of K-40, U-238 and Th-232, respectively.

2.5 Analysis of heavy metals by ICP-AES

Marine sediment was digested according to USEPA [11]. 1 g of dried sediments was digested in 10 mL of 8 M nitric acid over a hot plate at 92 – 95 °C for 10 min. After cooling, 5 mL of concentrated nitric acid was added with continuous heating for 30 min. Deionized water (2 mL) and 30% hydrogen peroxide (3 mL) were added; the digested sediment was then heated until bubbles disappeared. Hydrochloric acid...
and 2 mL of deionized water were then added; the mixture was heated for 10 min, then left to cool and filtered through Whatman paper no. 42. The solution was adjusted to 100 mL with deionized water.

Concentrations of heavy metals As, Cd, Cr, Cu, Hg, Ni, Pb and Zn were determined by a pre-calibrated ICP-AES (JY 2000, Jobin Yvon) with detection limits of 2.82, 0.32, 4.49, 0.002, 0.05, 0.12, 0.007 and 0.05 ppm, respectively.

Benthos samples were digested and analyzed in the same manner as the sediments, following classification of the benthic communities. All experiments were performed in three replications.

3. Results and discussion

3.1 Hydrographic parameters of water and sediment texture

The hydrographic parameters of water were presented in table 1. Water depth of the sample sites ranged from 1.7 - 7.8 m; the deepest site was located in an artificial channel of a commercial pier. The natural depth did normally not exceed 3 m at 1 km distance away from the coast line. The sandy sediment made the sea water highly transparent. The salinity was less than what was normally observed for sea water (35 ppt) due to dilution by fresh water from a water discharge close to the sea. Dissolved oxygen (DO) was slightly lower than the standard values (6.1 - 6.6 ppm at 20 - 30 °C, 1 atm, [12]) except at Si Racha pier (site 6), Si Racha Shipping area (site 7) and Leam Chabang pier (site 8), where the deep sea piers were located causing high water turbulence and a lower organic content (data not shown). The lowest DO was found at Si Racha Fisheries Research Station (site 5) and Bang Lamung (site 9). The former was a mussel farm area and the latter consisted of local fisheries and the beach area, where there were plenty of organic matters from fisheries and anthropogenic activities along the beach. The temperature showed less variation in the tropical area and along the coastal line.

The marine sediments at all sampling sites were characterized as sand using the soil texture triangle [8] since they contained more than 60% sand particles. Their sand particles content varied from 64.20 ± 12.33 - 94.73 ± 2.34%. The sediment samples collected nearby the beach (sites 2, 3, 4 and 10) contained more than 80% sand particles with small amounts of silt and clay. The sand and silt contained fewer amounts of fine particles and organic matters which allowed only small amounts of deposition and adsorption of radionuclides and heavy metals.

| Site | Depth (m) | Temp. (°C) | Transp. (m) | Salinity (ppt) | EC (mS/m) | DO (mg/L) |
|------|-----------|------------|-------------|----------------|-----------|-----------|
| 1    | 1.7       | 29.4       | 1.3         | 21.7           | 22.2      | 4.96      |
| 2    | 3.5       | 29.4       | 2.7         | 20.0           | 34.3      | 4.83      |
| 3    | 1.7       | 29.8       | 0.7         | 20.8           | 35.2      | 4.84      |
| 4    | 1.5       | 26.7       | 1.2         | 17.4           | 28.9      | 5.01      |
| 5    | 4.6       | 30.1       | 2.1         | 28.4           | 44.3      | 2.84      |
| 6    | 2.3       | 30.1       | 1.9         | 27.5           | 44.7      | 6.13      |
| 7    | 7.3       | 28.7       | 2.0         | 24.6           | 45.4      | 6.42      |
| 8    | 7.8       | 29.5       | 3.0         | 28.9           | 46.5      | 6.71      |
| 9    | 2.5       | 29.4       | 1.1         | 29.1           | 46.7      | 3.31      |
| 10   | 3.8       | 29.4       | 1.5         | 19.6           | 47.5      | 4.05      |

Mean±SD 3.67 ± 2.28 29.25 ± 0.98 1.75 ± 0.73 23.8 ± 4.42 39.57 ± 8.86 4.91 ± 1.27

Temp. = temperature; Transp. = transparency; EC = electrical conductivity; DO = dissolved oxygen.
Spatial and temporal variation of radionuclides in marine sediments

Naturally-occurring radionuclides including U-238, Th-238 and K-40 were found in marine sediment samples (table 2). The specific activity of U-238 was calculated from mean specific activities of Pb-214 (351.92 keV) and Bi-214 (609.31 keV); similarly, the specific activity of Th-232 was calculated from Tl-208 (583.00 keV) and Ac-228 (911.07 keV). The specific activity of K-40 was calculated from the photopeak at 1,460.81 keV. However, the photopeak of Cs-137 was not found in the marine sediments where its minimum detectable activity (MDA) was 2.46 Bq/kg.

UNSCEAR [13] reported the global mean specific activities of U-238, Th-232 and K-40 not exceeding 35, 30 and 400 Bq/kg, respectively. Mean specific activities of U-238, Th-232 and K-40 in sediments collected from Si Racha Fisheries Research Station (site 5) and Si Racha Shipping area (site 7) were less than the global mean values. Low specific activities of U-238 and Th-232 and high specific activity of K-40 were found at Wornapa beach (site 2) in three seasons. The mean specific activities of U-238, Th-232 and K-40 were highest in the rainy season and were also higher than the global mean values. In winter and summer, specific activities of U-238 and Th-232 were lower, while the specific activity of K-40 was much higher than the global mean values.

Due to the difference in precipitation of rainwater and the direction of Monsoon [14], the mean specific activities of the three selected radionuclides in summer and winter were higher than in rainy season except at Wastewater discharge, Bangpra (site 3). The accumulation of radionuclides in marine sediments was affected by the other factors such as geography, sand or clay beach, presence of piers, wastewater discharge sites, mussel farms and so on. The accumulation of radionuclides at Si Racha Fisheries Research Station (site 5) was lower than the other sampling sites because this area was covered with mussel farms. The high amount of calcium carbonate and the porosity of mussel shells [15] led to a high potential of adsorption resulting in low accumulation of radionuclides in the marine sediment.

The mean specific activities of U-238, Th-232 and K-40 from 10 sampling sites were provided in table 2. The values were higher than the global mean values reported by UNSCEAR (35, 30 and 400 Bq/kg, respectively).

The total activities of individual sampling sites in figure 2 indicated the lowest activity at Si Racha Fisheries Research Station (site 5) followed by Si Racha Shipping area (site 7). The total activities were highest at Wastewater discharge, Bangpra (site 3) in the rainy season. This area was located closed to the water discharge site where the strong water stream flowed through the sea causing the transportation of radionuclides from inland to the marine sediment. The radionuclides may be transferred together with the sediment by water runoff in the rainy season. The grains of clay and silt particles were very small with an enormous surface area and a net negative charge; hence, cations of radionuclides would easily be adsorbed to the surface of these particles with an electrostatic force. The specific activity of K-40 was approximately ten times the magnitude of U-238 and Th-232. Additionally, the ionic radius of K-40 (220 pm [16-17]) was approximately similar to the interlayer distance of non-expanding 2:1 silicate clay [18]; therefore, it was easily adsorbed to the sediment. The ionic radius of U-238 (175 pm) and Th-232 (180 pm [16-17]) were approximately 40 pm smaller than potassium; they could not fit into the interlayer distance of silicate clay. The expected adsorption or ion exchange at the outer surface of silicate clay, which was actually dominant to electrostatic interaction, was the only mechanism. The highest activity in the summer was found at Si Racha pier (site 6). In winter, the highest activities were found at Wornapa beach (site 2) and Bang Lamung (site 9). In addition to the large fraction of sand in these three sampling sites, the less adsorption should be found. Organic matters played an important role to enhance the adsorption by chemical bonding at their active sites (carboxylic, amino, phenolic and hydroxyl sites of organic matters). The sediment contained a small amount of organic matters (< 0.5%); as a result, it did not strongly affect the adsorption mechanism of radionuclides to the surface of the sediment. The large accumulation of radionuclides could be originated from a large amount of wastewater discharge to the sea.
Table 2. Spatial and temporal variation in specific activities of radionuclides in the marine sediments.

| Site | Nuclide | Specific activity (Bq/kg) | Mean ± SD |
|------|---------|--------------------------|-----------|
|      |         | Rainy season 2010 | Winter 2010 | Summer 2011 |
| 1    | $^{238}$U | 38           | 56           | 66         | 53±7 |
|      | $^{232}$Th | 63           | 86           | 92         | 80±7 |
|      | $^{40}$K  | 524          | 572          | 589        | 562±16 |
| 2    | $^{238}$U | 14           | 19           | 27         | 20±3  |
|      | $^{232}$Th | 14           | 19           | 23         | 19±2  |
|      | $^{40}$K  | 977          | 954          | 223        | 718±202 |
| 3    | $^{238}$U | 128          | 22           | 41         | 63.±27 |
|      | $^{232}$Th | 208          | 25           | 23         | 85±50 |
|      | $^{40}$K  | 986          | 649          | 628        | 754±95 |
| 4    | $^{238}$U | 66           | 73           | 30         | 56±11 |
|      | $^{232}$Th | 95           | 110          | 39         | 81±18 |
|      | $^{40}$K  | 443          | 556          | 481        | 493±27 |
| 5    | $^{238}$U | 18           | 20           | 20         | 19±0.5 |
|      | $^{232}$Th | 18           | 22           | 24         | 22±1  |
|      | $^{40}$K  | 99           | 120          | 132        | 117±8 |
| 6    | $^{238}$U | 24           | 26           | 33         | 29±6  |
|      | $^{232}$Th | 28           | 30           | 37         | 27±6  |
|      | $^{40}$K  | 151          | 153          | 184        | 494±14 |
| 7    | $^{238}$U | 24           | 26           | 33         | 27±2  |
|      | $^{232}$Th | 28           | 30           | 37         | 31±2  |
|      | $^{40}$K  | 151          | 153          | 184        | 163±9 |
| 8    | $^{238}$U | 37           | 45           | 41         | 41±2  |
|      | $^{232}$Th | 41           | 53           | 49         | 48±3  |
|      | $^{40}$K  | 287          | 381          | 366        | 345±24 |
| 9    | $^{238}$U | 37           | 69           | 42         | 49±8  |
|      | $^{232}$Th | 48           | 78           | 50         | 58±8  |
|      | $^{40}$K  | 547          | 817          | 700        | 688±64 |
| 10   | $^{238}$U | 45           | 40           | 75         | 54±9  |
|      | $^{232}$Th | 40           | 38           | 62         | 47±6  |
|      | $^{40}$K  | 409          | 379          | 401        | 396±7 |
| Mean ± SD | $^{238}$U | 44±10 | 41±6 | 39±6 |
|       | $^{232}$Th | 59±17 | 50±19 | 41±7 |
|       | $^{40}$K  | 463±94 | 484±83 | 472±81 |

Radiation hazards associated with radioactivity in the sediments were assessed from the measured specific activity as shown in table 3. The absorbed dose rate in air ranged from 27-112 (mean: 69 ± 8) nGy/h with some sites having the value higher than Thail and’s mean value (77 nGy/h [13]). Radium equivalent activity ($\text{Ra}_{\text{eq}}$) ranged from 59 – 243 (mean: 149 ± 19) Bq/kg, which was less than the limit value (370 Bq/kg [19]). External hazard index ($H_{\text{ex}}$) ranged from 0.2 – 0.7 (mean: 0.4 ± 0.05), which was less than 1 [20]. Finally, annual external effective dose rate ($\text{AED}_{\text{out}}$) ranged from 0.03 – 0.14 mSv/y (mean: 0.08 ± 0.01 mSv/y) which was less than the global mean (0.48 mSv/y; [13]). Furthermore, the average annual external effective dose rate was lower than the maximum permissible level (MPL) [13].
Figure 2. Temporal variation of total specific activities in the marine sediments.

Table 3. Risk index of natural radionuclides.

| Site | D (nGy/h) | Ra_{eq} (Bq/kg) | H_{ex} | AED_{out} (mSv/y) |
|------|-----------|-----------------|--------|------------------|
| 1    | 97        | 211             | 0.6    | 0.12             |
| 2    | 51        | 102             | 0.3    | 0.06             |
| 3    | 112       | 243             | 0.7    | 0.14             |
| 4    | 96        | 210             | 0.6    | 0.12             |
| 5    | 27        | 59              | 0.2    | 0.03             |
| 6    | 51        | 106             | 0.3    | 0.06             |
| 7    | 38        | 85              | 0.2    | 0.05             |
| 8    | 62        | 136             | 0.4    | 0.08             |
| 9    | 87        | 186             | 0.5    | 0.11             |
| 10   | 69        | 151             | 0.4    | 0.09             |
| Mean ± SD | 69 ± 8 | 149 ± 19 | 0.4 ± 0.05 | 0.08 ± 0.01 |

3.3 Concentrations of heavy metals in marine sediments

The high amounts of Fe (4,175 ± 183 and 8,307 ± 868 mg/kg) were found in rainy and winter seasons, respectively. The important sources of Fe were transportation and import/export goods containing iron. Zheng reported that a high concentration of metals would normally be found near the source [21]; for example, the high concentration of Zn (19,786 mg/kg) was found near the Huludao Zinc Plant; the largest zinc plants in Asia. In contrast, radionuclides could be found far away from their sources because they could accompany various kinds of media. Besides the atmospheric phase, they could accompany fallouts or gaseous-phase radionuclides such as tritium in titrated water vapor and iodine vapor. Wind speed and direction were the most critical factors to bring them far away from the source. Radionuclides could also mobilize in the aquatic phase; for example, Cs-137 was always found in marine ecology being far away from the source. Finally, both heavy metals and radionuclides would settle down and accumulate in the marine sediments. Manganese was used as an anti-corrosion coating for columns of docks and jetties [22]. A high concentration of Mn could be harmful to embryos of marine animals [23]. Lead was the third most abundant metal to be found in marine sediments particularly at the Sanctuary of Truth (site 10), Bang Lamung (site 9) and Si Racha pier (site 8) (260.69±8.42, 233.21±7.13 and 205.74±8.20 mg/kg, respectively) with average value of 154.28±26.68 mg/kg. It was probably carried
by wind or water stream from the Si Racha shipping area. Spraying of primer paints containing lead oxide may be the source of Pb in marine sediment.

In winter, Cr was the second most abundant metal at Si Racha Shipping area, Leam Chabang pier and the water discharge site, Bangpra (1,168.10 ± 126.44, 934.11 ± 32.48 and 692.23 ± 15.47 mg/kg, respectively). This was likely due to the fact that Cr was used as a metal coating to prevent oxide formation, a mixture of vanish oil and a primer in ship repairing process. Mn was the third most abundant at Fish dock and Si Racha pier (593.82±13.02 and 356.86±1.35 mg/kg).

The concentration of each metal in winter was lower than in rainy season except Pb which was probably originated from the shipyard and transported by the southwest monsoon (a seasonal rainy wind). However, the accumulation of metals in marine sediments was slightly low in the rainy season due to the dilution by rainwater.

Since the industrial estate and shipping activities spanned the area from Leam Chabang pier to the Sanctuary of Truth (site 10), contamination of heavy metals in the marine sediments was higher than other areas from Fish dock, Chonburi (site 1), to Si Racha Fishery Research Station (site 5). Fine particles with a large surface area and plenty of exchangeable sites were found leading to high accumulation of metals to their surface. From Fish dock, Chonburi, to Si Racha Fishery Research Station, sand particles dominated and the area was used for habitats and tourism resulting in the low concentration of metals.

In winter, the concentration of Pb reached its maximum followed by Hg, Cu, Cd, Ni, Zn and Cr. Similar to radionuclides, the highest level was found at Si Racha pier (site 6) followed by Wastewater discharge, Bangpra. The former might have originated from the source of contamination itself. The latter might have been originated from the source with a large amount of organic matters from water discharge, which enhanced the adsorption of metals, and also a large amount of clay and silt. Similar to Wastewater discharge, Bangpra (site 3), the coastal sediment consisted of a high percentage of silt and clay; the large amount of Pb originated from Bang Lamung (site 9) was found in coastal sediments collected from the Sanctuary of Truth (site 10).

3.4 Relation between benthos and heavy metals
Populations of benthos varied from 45 – 4,447 individuals/m² and the diversity indices ranged from 0 - 2.2. The diversity index reflected the relationship between species and number of individuals in each aquatic system. The highest diversity of benthos (15 species) was found at Wastewater discharge, Bangpra (site 3) followed by Wornapa beach (site 2); the Sanctuary of Truth (site 10); Fish dock, Chonburi (site 1), which comprised of 14, 13 and 12 species, respectively. The lowest diversity was found at Bang Lamung (site 9) where only 2 species were found. Annelida and Mollusca were found in each sampling site; these groups of organisms were more tolerant to high pollutant levels than other phyla. The others ware Arthropoda and Echinodermata. Some economic species including *Portunus pelagicus* and *Penaeus merguiensis* were also found. Fe, Mn, Pb and Cr in marine sediments were not significantly transported to benthos along the study sites, although these metals were highly accumulated to marine sediments.

Four phyla of benthos consisted of Annelida (16 species), Arthropoda (7 species), Mollusca (19 species) and Echinodermata (2 species); totally, 44 species were found among the ten sites. Polychaetes, shrimps, crabs, gastropod bivalves, sea cucumber and echinoderms were dominated at each station. The highly tolerant species were polychaetes (Annelida) and gastropods (Mollusca) with higher numbers of individuals (data not shown).

In this study, the radionuclide level in benthos was not measured since the numbers of benthos were not enough for radioactivity counting.

4. Conclusion
Besides the assessment of radionuclide accumulation in marine sediment along the Upper Gulf of Thailand by HPGe-gamma spectrometry, the natural radionuclides including U-238, Th-232 and K-40 were found with mean specific activities of 39 ± 6, 41 ± 7 and 472 ± 81 Bq/kg in summer; 44 ± 10, 59
and 463 ± 94 Bq/kg in rainy season, and 41 ± 6, 50 ± 9 and 484 ± 83 Bq/kg in winter, respectively. No contamination of the artificial radionuclide (Cs-137) was found. Accumulation of radionuclides in marine sediments depended on chemical properties of radionuclides, physical and chemical properties of the sediments: type of sediments, grain size and organic content, and geography: season, stream direction and flow rate. The radionuclide levels in marine sediments were higher than the global mean values reported by UNSCEAR. However, the radiation hazards associated with the radioactivity in the sediments were not expected.

Although no contamination of fission product including Cs-137 was found, the radionuclide level was very important since the sampling time was a few days before the Fukushima Daiichi disaster in 2011. The results under this study would be useful as a baseline data supporting the environmental radiation surveillance program of the country. Continuous monitoring after Fukushima disaster should be performed to trac any changes in the radioactivity levels and to ensure the radiation safety of the marine environment.

Metal contaminants in the marine sediments could be divided into two groups: less toxic (Fe and Mn) and highly toxic metals (Cd, Cu, Cr, Hg, Ni, Pb and Zn). For less toxic metals, Fe reached its highest concentration in both rainy and winter seasons, followed by Mn in the rainy season. Among toxic elements, Pb and Cr reached the highest in the rainy season (154.28±26.68 mg/kg) and in winter (578.16±126.74 mg/kg), respectively. The amount of metals was slightly higher in winter than in the rainy season due to water dilution, strong wind and waves in the rainy season. However, the amount of heavy metals did not exceed the standard metals contamination in marine sediments. A high contamination was found at the industrial and shipping areas.

Assessment of radionuclides and heavy metals along the Upper Gulf of Thailand was very important to address the concern about human risk to exposure. The benthic communities could be used as a biological index of a tolerance level guideline for primary producers and consumers. Continuous monitoring of toxic materials not only radionuclides and heavy metals but organic compounds (PAHs, VOCs, pesticides and others) was very important in marine surveillance and could be used as baseline data for management of marine ecology. The amount of toxic substances should be evaluated together with soil parameters: pH and organic matters, to determine the kinetic and thermodynamic parameters.

Acknowledgement
We would like to express our sincere thanks to Office of Atoms for Peace, Thailand, for the financial support. We thanked the Faculty of Science @ Si Racha and Si Racha Fisheries Research Station, Faculty of Fisheries, Kasetsart University, for laboratory facilities and ships as well as sampling facilities, respectively.

References
[1] Reddy M S, Bfsha S and Joshi H V 2005 India Chemosphere 61 1587-93
[2] Wang J and Chen C 2006 Biotechnol. Adv. 24 427-51
[3] Croudace I W and Cundy A B 1995 Environ. Sci. Technol. 29 1288-92
[4] Bothner M H, Buchholtz B M and Manheim F T 1998 Mar. Environ. Res. 45 125-55
[5] Lee S V and Cundy A B 2001 Estuar. Coast Shelf S. 53 619-36
[6] Finny B P and Huh C A 1989 Environ. Sci. Technol. 23 294-303
[7] Tessier A, Campbell P G C and Bisson M 1979 Anal. Chem. 51 844-51
[8] Boyd C E 1995 Bottom Soils, Sediment and Pond Aquaculture (New York: Chapman and Hall)
[9] IAEA 1989 Technical Report Series No. 295 Measurement of Radionuclides in Food and the Environment: a Guidebook (Vienna: IAEA)
[10] Veiga R, Sanches N, Anjos R M, Macario K, Iguatemy M, Aguiar J G, Santos A M A, Mosquera B, Bastos J, Carvalho C, Fiho M B and Umisedo N K 2006 Radiat. Meas. 41 189-96
[11] Edgell K 1989 USEPA Method Study 37 SW-846 Method 3050 Acid Digestion of Sediments, Sludges and Soils (Cincinnati: USEPA)
[12] USEPA 2012 *Method 3050B Acid Digestion of Sediments, Sludges, and Soils* (Washington, DC: USEPA)
[13] UNSCEAR 2000 *Sources and Effects of Ionizing Radiation 2000 Report to the General Assembly with Annex B: Exposure from Natural Source of Radiation* (New York: UNSCEAR)
[14] Wongsanit S 2010 *Gross Alpha and Beta Activities in Sea Water, Sediment and Plankton in the Coastal Ecosystem: the Upper Gulf of Thailand* (Bangkok: Kasetsart University)
[15] Tubtong C and Juijunjern R 2005 *Removal of Heavy Metals from Wastewater by Mussel Shell* (Bangkok: Srinakarinwirot University)
[16] Shannon R D and Prewit C T 1969 *Acta. Cryst.* B25 925-46
[17] Shannon R D 1976 *Acta. Cryst.* A23 751-61
[18] Essington M E 2003 *Soil and Water Chemistry, an Integrative Approach* (Boca Raton: CRP Press)
[19] OECD 1979 *Exposure to Radiation from the Natural Radioactivity in Building Materials Organization for economic Cooperation* (Paris: OECD Nuclear Energy Agency)
[20] El-Taher A and Madkour H A 2011 *Appl. Radiat. Isotopes* 69 550-658
[21] Zheng Y F, Wang Y J, Hou C L and Guan C M 2008 *Mar. Environ. Sci* 02 (in Chinese)
[22] Parthiban G T, Palaniswamy N and Sivan V 2009 *Anti-Corros. Methods Mater.* 56 78-83
[23] Columina M T, Domingo J L and Llobel J M 1996 *Veterinary and Human Toxicol.* 38 7-9