Risk Assessment of Radon Concentrations in Water of the Tapi River Nearby the Tapi Estuary of Bandon Bay, Muang District, Surat Thani Province

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ABSTRACT

This work aimed to study the physical characteristics and measurements of radon activity concentrations in water. In addition, the annual effective doses for water consumption obtained from the Tapi river nearby the Tapi estuary of Bandon Bay, Muang district, Surat Thani province, were estimated. The 90 water samples were collected from 15 sampling sites of the Tapi river nearby the industrial factories and riverside communities. The radon activity concentrations in water were measured using the RAD7 electronic radon detector. The average values of electrical conductivity, background radiation dose, temperature, and pH were 0.33±0.32 S/m, 0.62±0.22 mSv/y, 30.41±0.99 °C, and 7.01±0.27, respectively. The radon activity concentrations were in the range of 0.06–0.97 Bq/L, with an average value of 0.37±0.18 Bq/L for all measured samples in the study area. These results indicated that all measured radon concentrations were lower than the alternative maximum contaminant level for raw water (148 Bq/L) recommended by the U.S. EPA. To estimate the annual effective doses for water intake in infants, children, adults, and the weighted estimate of consumption, the results were significantly lower than the proposed safe limit of 100 μ Sv/y recommended by the WHO. This work could be applied for further study on the correspondence of radon variation and submarine groundwater discharge into the Bandon Bay through the Tapi river. Moreover, the correlations of radon concentration in consumption water, soils, sediment in the river, and indoor radon in dwellings nearby the riverside area will be necessary to confirm the health safety due to radon.

Keywords: radon in water, RAD7, annual effective dose, Tapi river, Bandon Bay

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Introduction

It's well known that radon (Rn-222) is a natural radioactive, colorless, odorless, and tasteless noble gas. Radon is formed by the decay of radium-226 (Ra-226), which originates from the decay of uranium-238 (U-238) found in soil, rock, and groundwater [1]. Moreover, the uranium-238 could be found in sand and sediment of sea beach areas [2-3]. The greatest exposure to radon occurred in the house [4]. Radon has a half-life $(t_{1/2})$ of 3.82 days which is enough to stay in the environmental atmosphere because of long-lived U-238 and Ra-226 radionuclides. It emits an alpha particle during decay which transforms into a series of solid, short-lived daughter products, of which polonium-218 (Po-218) and polonium-214 (Po-214). These daughter products will decay rapidly and also emit highenergy alpha particles. After inhalation of radon, the decay process will occur inside the lung, the emitted alpha particles may damage lung tissue, which can cause lung cancer in humans [1, 5]. The cause of radon up to 15% of lung cancer incidences worldwide was estimated by the World Health Organization (WHO) [4]. In the case of radon ingestion, the alpha particles emitted from radon dissolved in drinking water and its daughters could also be the risk of stomach cancer [6]. Radon could be generated from water distribution-system radium adsorbed iron pipe scale deposits [7]. Waterborne radon can lead to health risks by two pathways, which are radon inhalation due to the release of radon gas from water into indoor air, and the direct ingestion of radon in drinking water. The risk of waterborne radon was found about 89% of lung cancer caused by inhaling radon released from the water and 11% of stomach cancer caused by receiving contaminated radon in water [8]. However, the association between consumption of drinking water containing radon and the increased risk of stomach cancer was not confirmed by epidemiological studies. Many international organizations have recommended some regulations for radon concentrations in water. The maximum contamination level (MCL) for radon concentration in drinking water was recommended by the United States Environmental Protection Agency (U.S. EPA) in 1991. They proposed a maximum contaminant level (MCL) at the level of 11.1 Bq/L and radon in raw water not exceed the alternative maximum contaminant level (AMCL) of 148 Bq/L [9]. The WHO and the EU (European Union) Directive EC2013/51/EURATOM recommended that the safety limit for radon concentration in drinking water should not be higher than 100 Bq/L [10-11]. The United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR) suggested radon activity levels in water for human depletion in the range of 4-40 Bq/L [12]. Moreover, the WHO has determined the permitted level of 0.1 mSv/y (100 μ Sv/y) for an annual effective dose of radon in drinking water [13].

The radon activity concentrations in surface waters have been studied in many countries for a long time. Recently, radon concentrations in natural sources were measured for the study of radon activity distribution in potable water (see more details in Table 2). In most locations, the radon activities were found in the lower levels compared to the MCL and AMCL. Nevertheless, the average radon concentrations of some areas were found to be higher than the MCL, such as radon in potable water of Uttara Kannada district (Coastal region), Karnataka state, India [14], and radon in well water of the Namom district (Southern Thailand) [15]. Regarding the health risks for radon consumption, the radon

concentrations in water obtained from natural sources should be measured for risk assessment of radon in potable water. It is an important step in preventing radon exposure.

Tapi river is located nearby the Tapi estuary of Bandon Bay, Muang district, Surat Thani province. There are nearby the industrial factories and riverside communities where may compose of radionuclides contamination in water released from the production process of industrial factories as well as municipal solid waste left from the riverside communities. Currently, the Tapi river has been used for consumption and tourism recreation. The Tapi river is a source of raw water for producing tap water. There are also factories located on both sides of the Tapi river that use water for the consumption of large quantities. Reasonably, the radon activity concentrations in the Tapi river should be studied for health risk assessment due to radon.

In this work, the physical characteristics and radon concentrations in the Tapi river were analyzed. Additionally, the annual effective doses for water consumption obtained from the Tapi river were estimated. However, Thailand has not set any reference level for radon concentration in consumption water. Thus, the measured radon concentrations in water in this work will be compared to the AMCL recommended by the U.S. EPA as well as the annual effective dose due to radon ingestion will be compared to the safety limit recommended by the WHO.

Materials and Methods

Water sample collection and preparation

The study area of the Tapi river, in Muang district, Surat Thani province, is located between north latitudes ranged from 9.168329 °N (1013481 m) to 9.169446 °N (1013608 m) and east longitudes ranged from 99.350845 °E (538545 m) to 99.381702 °E (541935 m). There is nearby the Tapi estuary of Bandon Bay, where is a boundary line between freshwater and seawater in the Gulf of Thailand. Most people who live in the riverside of the study area and the industrial factories located nearby the Tapi river use water for consumption. Thus, radon exposure may happen during water consumption. In this study, the water sampling area was selected nearby the industrial factories and riverside communities because the radionuclides contamination in the Tapi river could be released from the production process of industrial factories and municipal solid waste might be left from the riverside communities.

The water samples were collected from 15 sampling sites. The samples from each sampling site were collected in 3 samples and collected repeatedly two times amount to 90 samples for all collections. The duration time for sample collection was around 01.00-03.00 P.M. because of the slightly clouded and calm wind waves of July to August 2020. The long-tailed boat was used for sample collections from each position, with a distance about 15-20 m away from the coast because of the constant shore wave that was suitable for water sampling.

The handheld GARMIN GPS with the model of eTrex 20x was used for positioning sampling location using the Universal Transverse Mercator (UTM) coordinate system identified by B1-B15 shown in Figure 1 and Table 1. The water samples were collected at a water depth of about 1 m from the water surface using a 250 mL collection vial. The vial cap would be tightened while still under the water to make sure that there were no bubbles in the collection vial. The vial was taken from the water and dried. Then, the cap was sealed again with parafilm against the decay of radon gas from a vial cap. The collected water samples were then brought to the laboratory for radon concentration measurement using the RAD7 electronic radon detector.

The physical characteristic study

To study the physical characteristics of coastal waters in the selected area, the background radiation dose (BG), electrical conductivity (EC), temperature (T), and pH were measured. Background radiation doses were measured at sampling sites using the Ranger Survey Meter with the model CE0197. The unit of background radiation dose was displayed in microsieverts per hour $(\mu Sv/h)$ and then transformed into mSv/y to a comparison of the world average natural radiation level of 2.4 mSv/y or in the range of 1-10 mSv/y reported by the UNSCEAR [1]. The electrical conductivity and temperature of water samples were also measured at the sampling sites using the Eutech Instruments with the model of Cyberscan COND 610 displayed in the units of Siemens per meter (S/m) and degree Celcius ($\rm{^o}C$), respectively. The Mettler Toledo FiveEasy pH bench meter was used for measuring pH levels in water samples after measuring radon concentrations in the laboratory.

Figure 1 The geological map of the Tapi river nearby the Tapi estuary of Nabdon Bay was taken from Google Earth, with the geographic coordinate system codes of the sampling sites identified by B1-B15.

The experimental method for measurement of radon concentration in water

In this section, the method for measuring radon concentration was described. The RAD7 electronic radon detector manufactured by Durridge Company Inc. (USA) connected to RAD H_2O accessory, was prepared to measure radon concentration in water samples shown in Figure 2.

Before start measuring, the RAD7 must be free of radon, and dry. Thus, it should be purged around ten minutes until the relative humidity inside the RAD7 device would stay below 10% for the entire 30 minutes of the measurement. If the RAD7 has not been used for some time, it will take longer to dry it out, perhaps as much as 30 minutes of purging, or even more. For measuring radon concentrations, the pump will run for four cycles with five minutes per cycle. At the end of the run, 30 minutes after the start, the RAD7 printed out a summary, showing the average radon reading from the four cycles counted, a bar chart of the four readings, and a cumulative spectrum. Additionally, RAD H₂O data can be graphed using DURRIDGE's CAPTURE software. All data, except the spectrum, was also stored in memory and could be printed or downloaded to a PC for data analysis [16].

Figure 2 The system setup for radon measuring in water samples using RAD7 electronic radon detector connected to RAD-H₂O accessory.

Analysis of radon concentrations in water

Usually, radon concentrations in water samples will decline due to radioactive decay because It was taken some time from sampling to analysis. Thus, the corrected radon concentrations will be calculated back from the measured time to the sampling time using the following equation 1 [17-18].

$$
C_{\scriptscriptstyle Rn} = C_{\scriptscriptstyle Rn-0} e^{-\lambda t} \tag{1}
$$

where C_{Rn} stands for the measured radon concentration, C_{Rn-0} represents the initial radon concentration calculated after the decay correction, λ is the decay constant for radon (2.1x10⁻⁶ 1/s or dps: disintegration per second) [19] and t represents the time elapsed since sample collection (s). The unit of corrected radon concentrations expressed in $Bq/m³$ will be converted to becquerels per liter (Bq/L) for standard comparison. The average radon concentrations (\bar{x}) and standard deviation (SD) of water samples collected from each position as well as the overall average radon concentration for all samples were analyzed. For the statistical hypothesis testing, the independent sample t-Test and One-way ANOVA were used for average comparisons of two groups and more than two groups of samples, respectively. The average radon concentrations were compared to the alternative maximum contaminant level for raw water (148 Bq/L) recommended by the U.S. EPA [9].

Estimation of annual effective doses for water consumption

The radiation doses to the stomach due to radon exposure in water were estimated by calculation of the annual effective dose due to the ingestion of radon for individuals of different age groups using the following relation by [1] (equation 2).

$$
Annual effective dose(Sv/y) = C_{\text{Rn}-0} \times C_{\text{w}} \times DCF
$$
\n(2)

where C_{Rn-0} represents radon activity concentration in water at the sampling time (Bq/L), C_w is the annual water consumption of tap water ingestion estimated in the UNSCEAR 1993 report [20] to be 100, 75, 50, and 60 L/y by infants, children, adults, and the weighted estimate of consumption, respectively, and the effective dose conversion factor (DCF) of radon intake by ingestion is $3.5x10^{-9}$ Sv/Bq [1]. The estimated values received from equation (2) would be compared to the safe limit for water consumption recommended by the WHO at the level of 0.1 mSv/y (100 μ Sv/y) [13].

Results and Discussion

The physical characteristics of water in the study area

The relations of electrical conductivity (EC) and background radiation dose (BG) in sampling sites were shown in Figure 3.

Figure 3 (a) showed the overall average EC value of 0.33 ± 0.32 S/m $(3.341\pm3.241 \text{ }\mu\text{S/cm})$. The average EC values of water in some sampling sites (B1-B5) were lower than the WHO standard value for drinking water (400 μ S/cm) [21], within the range of 0.034 \pm 0.004 to 0.039 \pm 0.005 S/m (339 \pm 41 to 390 \pm 46 μ S/cm). However, the average EC values of B1 to B7 sampling sites were not exceeded the guideline value of freshwater set by the WHO (1,500 μ S/cm), whereas the average EC in the sampling sites of B8-B15 were found in higher levels than the guideline value within the range of 0.20±0.03 to 0.91 ± 0.02 S/m (2,004 \pm 300 to 9,118 \pm 188 μ S/cm). The maximum average of EC value was found at the sampling site of B9 located in the nearest position to the Bandon Bay, whereas the minimum average of EC value was found at the B3 sampling site located at a great distance from Bandon Bay. The maximum and minimum values of EC in the study area were significantly different (Independent samples t-Test, two-tailed, $p < 0.05$). These results indicated that the sampling sites within the higher levels of EC values might be due to the location nearby the Tapi estuary of Bandon Bay. To consider the BG values of water in the sampling sites, Figure 3 (b) showed that the average BG values were in the range of 0.35±0.25 to 0.86±0.12 mSv/y with an overall average BG value of 0.62±0.22 mSv/y. These results indicated that BG values for all sampling sites did not exceed the world average BG natural radiation level of 2.4 mSv/y reported by the UNSCEAR [1]. However, the difference in average BG values for all sampling sites was not statistically significant (One-way ANOVA, $p > 0.05$). To analyze the relations of temperature (T) and pH levels of water in sampling sites, the results were shown in Figure 4.

Figure 3 The relations of electrical conductivity (EC) of water and background radiation dose (BG) in different sampling sites.

The average temperatures of water in different sampling positions were found within the range of 29.70 \pm 1.41 to 30.90 \pm 1.06 °C with an overall average of 30.41 \pm 0.99 °C (Figure 4 (a)). These results indicated that the measured temperatures for all samples were in the range of the safe limit at 30 $^{\circ}$ C [21]. To consider the pH of water in the study, the results showed that the pH levels varied from 6.69 \pm 0.22 to 7.79 \pm 0.05 with an overall average of 7.01 \pm 0.27 (Figure 4 (b)) within the range

of 6.5-8.5 for the safe limit [21]. Comparison of the average temperatures and pH levels between sampling positions were not statistically significant differences (One-way ANOVA, p > 0.05). However, the temperature and pH levels of water in the study area corresponded to other reports for river water nearby the factories and urban areas [22-25].

Figure 4 The relations of temperatures (a) and pH levels (b) of water in different sampling sites.

Radon concentrations in water and the annual effective doses for water consumption

Table 1 summarized the statistical analysis of radon concentrations in water and annual effective doses for water ingestion intake in different age groups in the investigated study. The positioning sampling locations were used the Universal Transverse Mercator (UTM) coordinate system identified by B1-B15. The number of samples (n), the maximum (Max), and the minimum (Min) values as well as the average radon concentration (\bar{x}) and standard deviation (SD) were used for the descriptive statistics and comparison of means.

Total

B6 539895 1014675 6 0.12 0.06 0.09±0.03 0.03±0.01 0.02±0.01 0.02±0.01 0.02±0.01 B7 540124 1014927 6 0.30 0.21 0.27±0.03 0.09±0.01 0.07±0.01 0.05±0.01 0.06±0.01 B8 540452 1015210 6 0.36 0.27 0.31±0.04 0.11±0.01 0.08±0.01 0.05±0.01 0.07±0.01 B9 541385 1015676 6 0.30 0.21 0.26±0.03 0.09±0.01 0.07±0.01 0.05±0.01 0.05±0.01 B10 540957 1014821 6 0.36 0.24 0.28±0.04 0.10±0.02 0.07±0.01 0.05±0.01 0.06±0.01 B11 541165 1014708 6 0.52 0.37 0.43±0.06 0.15±0.02 0.11±0.02 0.07±0.01 0.09±0.01 B12 541371 1014547 6 0.20 0.12 0.16±0.03 0.06±0.01 0.04±0.01 0.03±0.01 0.03±0.01 B13 541549 1014209 6 0.42 0.30 0.36±0.04 0.13±0.02 0.09±0.01 0.06±0.01 0.08±0.01 B14 541888 1013528 6 0.41 0.29 0.34±0.05 0.12±0.02 0.09±0.01 0.06±0.01 0.07±0.01 B15 541935 1013608 6 0.47 0.35 0.40±0.05 0.14±0.02 0.10±0.01 0.07±0.01 0.08±0.01

Table 1 Radon concentrations in water and annual effective dose for water consumption of different age groups in the study area.

Radon concentrations in 90 water samples from 15 different locations in the Tapi river were found to vary from 0.06 to 0.97 Bq/L with an overall average of 0.37±0.18 Bq/L. The maximum and the minimum radon levels were found at B2 and B6, respectively. These results indicated that radon concentrations in water in the investigated study were lower than the alternative maximum contaminant level for raw water (148 Bq/L) recommended by the U.S. EPA [9]. To consider the average annual effective doses for water consumption in infants (100 L/y), children (75 L/y), and adults (50 L/y), the results were found within the ranges of $0.03\pm0.01 - 0.30\pm0.03$, $0.02\pm0.01 - 0.23\pm0.02$, and 0.02 ± 0.01 – 0.15 ± 0.01 μ Sv/y, respectively, with the overall average annual effective doses in infants, children, and adults were 0.13 ± 0.06 , 0.10 ± 0.05 , and 0.06 ± 0.03 $\mu\text{Sv/v}$, respectively. Moreover, the total annual effective doses for the weighted estimate of consumption (60 L/y) were found within the range of $0.02\pm0.01 - 0.18\pm0.02$ μ Sv/y, with an overall average of 0.08 ± 0.04 μ Sv/y. Additionally, the annual effective doses for water consumption of different age groups could be confirmed that all levels were significantly lower than the safe limit for water consumption recommended by the WHO at the level of $100 \mu Sv/y$ [13].

Total 90 0.97 0.06 0.37±0.18 0.13±0.06 0.10±0.05 0.06±0.03 0.08±0.04

Sampling pos

Data in Table 1 were further analyzed into the bar charts shown in Figure 5. These results indicated that the radon concentrations in water of the B2-B4 sampling sites located in the factories nearby the industrial factories were found in significantly higher levels than the other sampling positions located nearby the riverside communities. These may be due to the influence of radionuclides contamination released from the production process of industrial factories [26-27]. Reasonably, the maximum average radon concentration was found in the level of 0.86±0.07 Bq/L at the B2 sampling position, while the minimum average radon concentration was 0.09±0.03 Bq/L at the B6 sampling position (Figure 5 (a) and see also Figure 1), which showed the statistically significant difference (Independent samples t-Test, two-tailed, $p<0.05$). Moreover, the consideration of the relationship between the EC values and the concentrations of radon in water, the data could be divided into two groups that were the low EC $(B1-B7)$ and the high EC $(B8-B15)$ zones (see also Figure 3(a)) The average activity radon concentration of the low EC zone (0.33±0.20 Bq/L) was significantly higher than the concentration in the high EC zone $(0.24 \pm 0.07 \text{ Bq/L})$ at 0.05 level (Independent samples t-Test, two-tailed, $p<0.05$). These indicated that the decrease of radon concentration may be due to the influence of surface water and the increase of EC may be due to the influence of seawater [28].

To consider the annual effective doses for water consumption in Figure 5 (b), the results showed the statistically significant difference for comparison of all average values in different age groups at 0.05 level (One-way ANOVA, p<0.05). The results showed that the maximum average annual effective doses for water consumption in infants, children, adults, and the weighted estimate of consumption analyzed from the B2 sampling position were 0.30 ± 0.03 , 0.23 ± 0.02 , 0.15 ± 0.01 and 0.18 ± 0.02 , respectively. While the minimum average annual effective doses for water consumption in infants, children, adults, and the weighted estimate of consumption analyzed from the B6 sampling position were found at the levels of 0.03 \pm 0.01, 0.02 \pm 0.01, 0.02 \pm 0.01, and 0.02 \pm 0.01 μ Sv/y, respectively. These results indicated that the decreasing trend of annual effective doses with the increase in age and decrease in water consumption rates. Additionally, the annual effective doses were proportional to the radon concentrations in water [1, 17].

Although some physical properties of water, radon concentrations in the water of the Tapi River, and the estimated annual effective doses for water consumption in this study were found much below the safety levels, all other parameters were indicators of water quality should be taken into account.

Figure 5 Radon concentrations in coastal waters and annual effective doses for water consumption of different age groups in the sampling sites.

Comparison of radon concentrations in surface waters for various types of water

Comparison of radon concentrations in surface waters for various types of water obtained from natural sources for different locations across the globe and the present study was showed in Table 2.

Table 2 Comparison of radon concentrations in surface waters for various types of water.

The concentrations of radon obtained in the present study were corresponding to the reports of river water obtained from Cauvery River, India, Khasa River, Kirkuk City, Iraq, River Hilla, Iraq, Da-Chia River, and Gaomei Wetland, Taiwan, whereas the radon levels obtained from river water of the Southwest coastal region of Peninsular Malaysia and Uttara Kannada district, India were in the higher levels than this work. Since this study site is located nearby the Tapi estuary of Bandon Bay, where is close to the coast of the Gulf of Thailand, radon concentrations in this study could also be compared to radon levels in the coastal waters. The radon concentrations in water of the Tapi river were also ranged approximately to the radon levels in coastal waters of Map Ta Phut, Rayong, Thailand [26] and Coast of Xiangshan, Zhejiang, China [35].

Moreover, radon concentrations in river water and coastal waters have been found in low levels compared to well water. These may be due to the aeration of radon to the atmosphere [26-27, 39-41]. Additionally, the lack of major contact with radon emanating mineral materials may be another effect of lower radon concentration in river water [40].

Conclusions

The average value of electrical conductivity was found at the level of 0.33 ± 0.32 S/m $(3341\pm3241 \,\mu\text{S/cm})$, which was higher than the guideline value of freshwater set by the WHO (1500) S/cm) [21]. However, 33 percent of sampling sites were lower than the standard value for drinking water (400 μ S/cm) recommended by the WHO [21], within the range of 0.034 \pm 0.004 to 0.039 \pm 0.005 S/m (339 ±41 to 390 ±47 $\mu S/cm$) obtained from the sampling site located at a great distance from Bandon Bay. These results could be concluded that high levels of EC values obtained from the sampling sites located closer to the Tapi estuary of Bandon Bay, the Gulf of Thailand were not recommended for water consumption. The average background radiation dose was found at the level of 0.62±0.22 mSv/y, which did not exceed the world average background natural radiation (2.4 mSv/y), reported by the UNSCEAR [1]. In addition, the overall average temperature and pH were 30.41 ± 0.99 ^oC, and 7.01 \pm 0.27, respectively, which were within the ranges of the safe limit of temperature (30 $^{\circ}$ C), and pH level (6.5-8.5) for drinking water recommended by the WHO [21]. The overall measured radon concentrations levels in various sampling sites obtained from the Tapi river were found in low and safe for water consumption; the concentrations of radon ranged from 0.06 to 0.97 Bq/L, with an average value of 0.37±0.18 Bq/L. The results indicated that the radon concentrations for all samples were very much lower than the alternative maximum contaminant level for raw water (148 Bq/L) recommended by the U.S. EPA [9]. In addition, the estimated annual effective doses for water consumption in infants, children, adults, and the weighted estimate of consumption were found within 0.13 ± 0.06 , 0.10 ± 0.05 , 0.06 ± 0.03 , and 0.08 ± 0.04 μ Sv/y, respectively, which indicated that the annual effective doses increased when a decrease in age and an increase in water consumption rate. These values were very much lower than the safe limit of $100 \mu Sv/y$ for water consumption recommended by the WHO [13]. Although the radon concentrations in water and the annual effective doses for water consumption in the study area were in extremely low values, It was still necessary to study because nobody could know the concentration of radon in water without measurement.

The data results of this study will be useful in drawing up the regulations for radiation protection from natural sources. The correlation between radon concentrations in consumption water, soils, sediment, suspended sediment in the river, and indoor radon in dwellings in the riverside area should be further studied for safety confirmation. Moreover, radon could be a good natural tracer of groundwater flow into the coastal waters [26, 42-43]. Additionally, this work could be applied for the study of the seasonal variation of radon concentration in water monitoring to study the effect of hydrological conditions [44-45].

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