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Radon and Thoron Exhalation Rates from Surface Soil of Bangka - Belitung Islands, Indonesia

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Abstract - Radon and thoron exhalation rate from soil is one of the most important factors that can influence the radioactivity level in the environment. Radon and thoron gases are produced by the decay of the radioactive elements those are radium and thorium in the soil, where its concentration depends on the soil conditions and the local geological background. In this paper, the results of radon and thoron exhalation rate measurements from surface soil of Bangka Belitung Islands at thirty six measurement sites are presented. Exhalation rates of radon and thoron were measured by using an accumulation chamber equipped with a solid-state alpha particle detector. Furthermore, the correlations between radon and thoron exhalation rates with their parent nuclide (^{226}Ra and ^{232}Th) concentrations in collected soil samples from the same locations were also evaluated. The result of the measurement shows that mostly the distribution of radon and thoron is similar to ^{226}Ra and ^{232}Th, eventhough it was not a good correlation between radon and thoron exhalation rate with their parent activity concentrations (^{226}Ra and ^{232}Th) due to the environmental factors that can influence the radon and thoron mobilities in the soil. In comparison to a world average, Bangka Belitung Islands have the ^{222}Rn and ^{220}Rn exhalation rates higher than the world average value for the regions with normal background radiation.

Keywords: radon, thoron, exhalation rate, soil, Bangka-Belitung

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Introduction

Radon (^{222}Rn) and thoron (^{220}Rn) are radioactive gases produced by the decay of ^{226}Ra and ^{224}Ra, which are themselves the decay products of ^{238}U and ^{232}Th series in the ground, respectively. ^{222}Rn and ^{220}Rn decay with the emission of alpha particles and produce daughter nuclei - polonium (^{218}Po, ^{216}Po, ^{214}Po, ^{212}Po), lead (^{214}Pb, ^{212}Pb, ^{210}Pb), and bismuth (^{214}Bi, ^{212}Bi,^{210}Bi). These daughter nuclei emit alpha or beta particles. ^{222}Rn has a half-life of 3.825 days and is an alpha emitter; ^{220}Rn has a half-life of 55.6 s and is also an alpha emitter (Figure 1) (Porstendorfer, 1994; Ramachandan and Sathish, 2011). ^{222}Rn having an atomic number of 86, is the heaviest member of the rare gas group (~ 100 times heavier than hydrogen and ~ 7.5 times heavier than air). ^{220}Rn is an isotope of radon, that has an atomic number of 86, and mass number of 220. The main characteristic of ^{222}Rn and ^{220}Rn among the other natural radioactive elements is the fact that their behaviour is chemically inert (noble gases), not affected by chemical processes. The ^{222}Rn and ^{220}Rn are free to move through soil pores and rock fractures; then to escape into the atmosphere. ^{222}Rn and/or ^{220}Rn exhaled from the earth surface into the free atmosphere is rapidly dispersed and diluted by

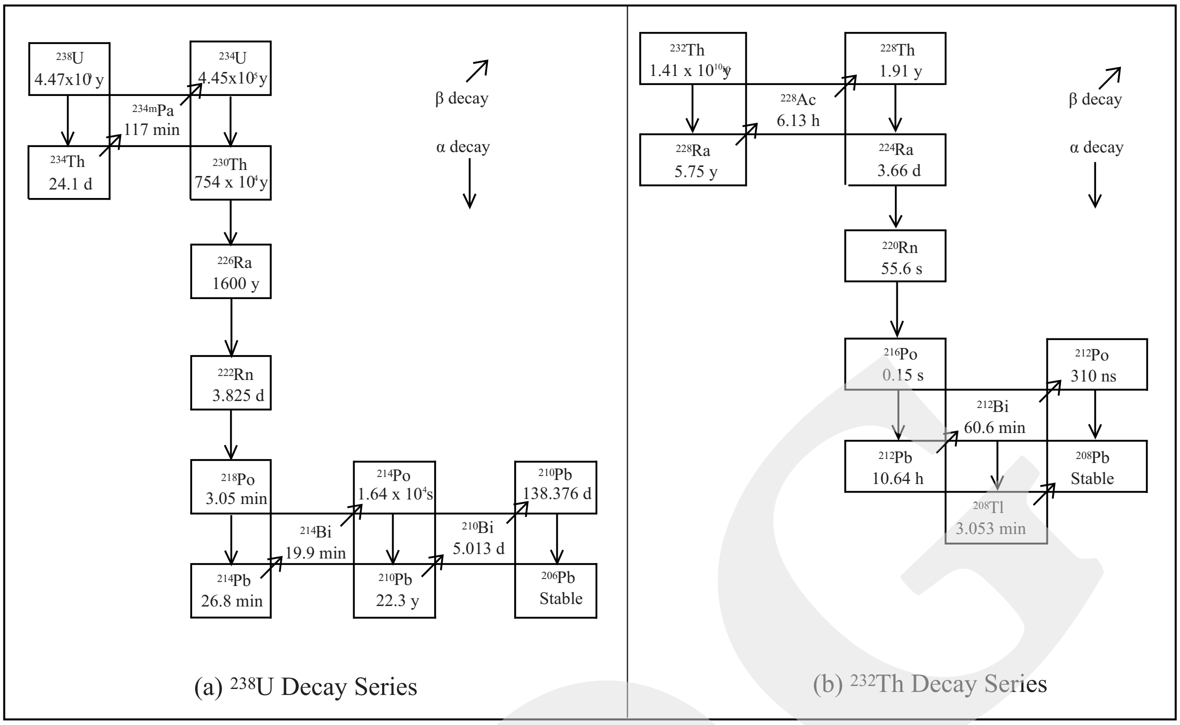

Figure 1. Decay Series of ^{238}U (a), and ^{232}Th (b) (HASL-300, 1997) .

natural convection and turbulence (Mudd, 2008; Hassan *et al.,* 2011).

^{222}Rn and ^{220}Rn in the ambient air depend on the soil conditions and the local geological background. ^{222}Rn and ^{220}Rn emanate mainly from the earth surface through the gap in soil to the atmosphere. ^{222}Rn and ^{220}Rn gases enter the house from various gaps in walls and open windows or doors. They decay producing isotopes of polonium (^{218}Po, ^{216}Po, ^{214}Po, ^{212}Po), lead (^{214}Pb, ^{212}Pb, ^{210}Pb), and bismuth (^{214}Bi, ^{212}Bi, ^{210}Bi) which are heavy metals chemically very active, that may exist briefly as ions and/or free atoms before forming molecules in a condensed phase or attached to airborne dust particles, forming radioactive aerosols. This fraction may be inhaled and deposited in the respiratory tract, in which they release all their α-emissions. It is, therefore, important to know the ^{222}Rn and ^{220}Rn exhalation, and these information are useful for presumption of a high ^{222}Rn-^{220}Rn concentration area.

Bangka and Belitung Islands have the geological potential of mineral resources, especially tin, with accessory minerals consisting of monazite, zircon, xenotim, ilmenite, magnetite, and pyrite spreading in almost all regions. Bangka Belitung Islands are known as tin producer places which form a part of Southeast Asia Tin Belt, the richest tin belt in the world which stretches along South China - Thailand - Myanmar - Malaysia to Indonesia (Schwartz *et al*., 1995). Meanwhile, there are some other mineral resources like: quartz sand, building construction sand, kaolin, granite, clay, and mountain stone. The geology of Bangka Belitung is structured by granites (hard stone). Itis generally covered by klabat granite which are devided into three catagories, *i.e.* biotite granite, granodiorite, and gneissic granite. The soil derived from granite will have a higher radioactivity than the soil from the other rock types (Saleh and Ramli, 2013: Rani and Singh, 2005).

Generally, when the ^{226}Ra and ^{232}Th are high, the exhalation rates of ^{222}Rn and ^{220}Rn at that site are a relatively high too. Therefore, this study was conducted with the ^{222}Rn and ^{220}Rn exhalation rates from the ground surface in Bangka-Belitung. The objective of this work is to determine ^{222}Rn and ^{220}Rn exhalation rates from the soil surfaces of Bangka – Belitung Islands and to evaluate the correlation with their parent radionuclides (^{226}Ra and ^{232}Th). The study will help in understanding the status of indoor and outdoor ^{222}Rn, ^{220}Rn and the status of the exhalation of these gases from soil in Bangka-Belitung. This type of study has never been done before in Bangka Belitung Islands.

Bangka-Belitung Islands are located at 104° 50' - 109° 30' E and 0° 50' - 4° 10' S which lie in east of Sumatra, northeast of South Sumatra Province (Figure 2).

Material and Methods

Material

The field study was performed on thirty six sampling sites where twenty eight sites were at Bangka Island and ten sites were at Belitung Island. Geographical coordinates of the sampling points were determined using GPS Map 60CHx manufactured by Garmin. After finding the measuring site, any grass, gravel, and roots were removed to perform the measurements of ^{222}Rn and ^{220}Rn exhalation rates. Then, at least 2 - 3 kg of soil was collected at each point using shovel and scoop. At a collection point the soil sample was wrapped in black plastic bag and then taken to the laboratory.

Measurement of ^{222}Rn and ^{220}Rn Exhalation Rates from Surface Soil

In this study, the method for ^{222}Rn and ^{220}Rn exhalation rate measurement is based on small accumulation chambers connected to a continuous Radon Gas Monitor, model RAD7 (Figure 3), produced by Durridge Company Inc. (2010), equipped with a solid state alpha detector (RAD7, Durridge Co. Inc., Bedford, MA, USA). The accumulation chamber made of stainless steel as ^{222}Rn and ^{220}Rn accumulation chambers is connected to the RAD7 detector by vinyl tubing with a gas-drying unit filled with a desiccant ($CaSO_4$ with 3% $CaCl_2$) to maintain the relative humidity at <10% within the measurement system. The system is a closed loop in which the gas circulates continuously.

The accumulation chamber was placed on the ground surface from its open side, and its surroundings were covered to prevent any air exchange with environment. Tamping down the soil around the chamber is to prevent the leakage of fresh air into the sample acquisition path or down the outside of the chamber to sampling point. Following the localization of the measured points using GPS devices and clearing the rel-

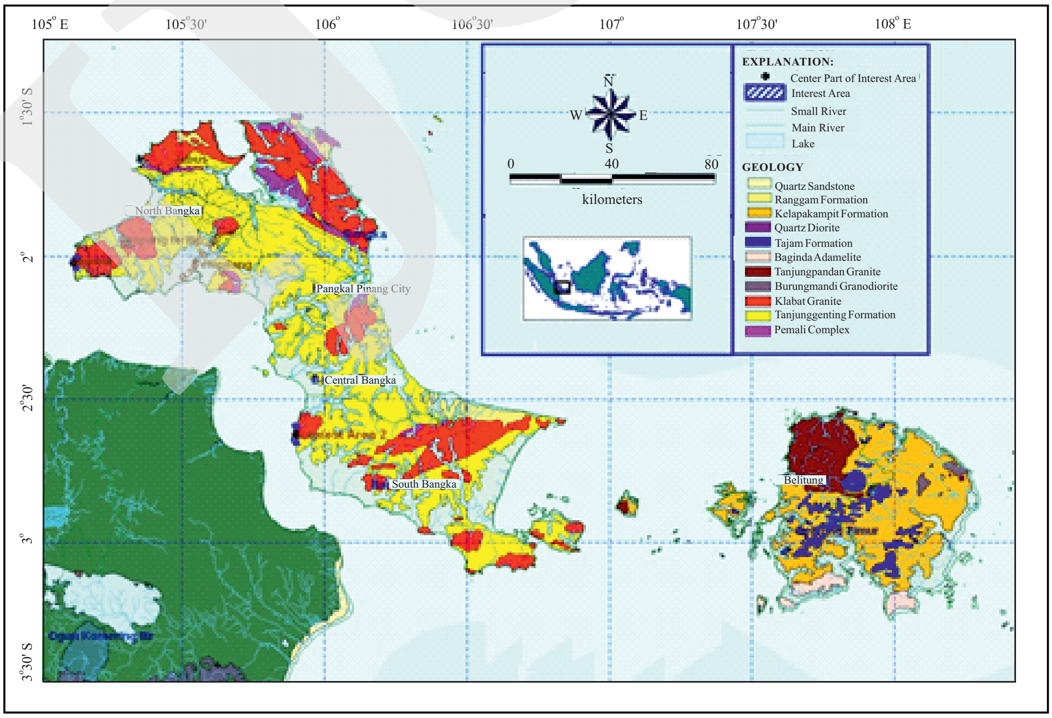

Figure 2. The geology map of Bangka Belitung (IAEA, 2011).

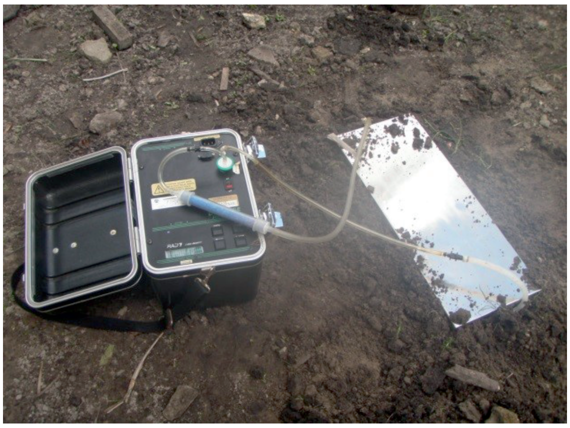

Figure 3. The RAD7 system and accumulation chamber.

evant surface of any grass, gravel, and plant roots, the ^{222}Rn and ^{220}Rn exhalation rate was measured. It is noteworthy that the environmental parameters such as temperature, pressure, and relative humidity were recorded by the device during the period of measuring each site.

The changes in the ^{222}Rn and ^{220}Rn concentration in the chamber were used as a function of time to estimate the exhalation rate from the ground surface. The concentration of ^{222}Rn and ^{220}Rn exhaled from the sample increases exponentially until radioactive secular equilibrium is reached. The exhalation rate (E_0) from the sample can be calculated with equation (Tuccimei *et al*., 2006; Hassan *et al*., 2011):

$$E_0 = \frac{Cx\lambda V}{S(1-e^{-\lambda t})} \quad \text{.....................................(1)}$$

where:

C is the net concentration (exhaled radon/thoron less the background concentration) at accumulation time t (Bqm^{-3}),

λ is the decay constant (s^{-1}),

V is the effective air volume (m^3), and

S is the sample surface area (m^2).

Measurement of ^{226}Ra and ^{232}Th in Surface Soil

The measurement of ^{226}Ra and ^{232}Th in the soil samples collected from the same site with ^{222}Rn and ^{220}Rn exhalation rates were carried out at a laboratory by using ORTEC P-type coaxial high purity Germanium (HPGe) detector with a relative efficiency of 60% and a resolution of 1.95 keV (full width at half maximum) for the peak of 1,33 keV. In the laboratory, the soil samples were dried in an oven at a temperature of 105^{0}C to a constant weight to remove any available moisture. After being dried, the samples were crushed and sieved with a mesh having holes each of diameter of 2 mm in order to remove organic materials, stones, and lumps. Afterwards, the homogenized samples were packed to fill 1 liter marinelli beakers. The marinelli beakers were carefully sealed in order to prevent trapped radon gas from escape and allowed to stand for at least four weeks for secular equilibrium to be established between the long-lived parent nuclides of ^{226}Ra and ^{232}Th, and their short-lived daughters before measurement.

The gamma energy peaks 352 keV of ^{214}Pb and 609.31 keV of ^{214}Bi were used to determine ^{226}Ra. The gamma energy peaks of 238.6 keV from ^{212}Pb, 911.2 and 969 keV gamma energy peak from ^{228}Ac and 583 keV gamma energy peak from ^{208}Tl were used to determine the ^{232}Th. The activity concentrations (A) of ^{226}Ra and ^{232}Th in Bq kg^{-1} for the samples were determined using the following expression (Knoll, 2000; Syarbaini *et al*., 2014):

$$A = \frac{N_e}{\varepsilon_f P_\gamma t_C M} \quad \text{..(2)}$$

where:

N_e = net counts of a peak at energy E,

ε_f = the counting efficiency of the detector system at energy E,

P_γ = the gamma ray emission probability (gamma yield) at energy E,

t_c = sample counting time,

M = mass of sample (kg).

Result and Discussion

The measurement result of ^{222}Rn and thoron exhalation rates is shown in Table 1. It can be seen in Table 1 that ^{222}Rn and ^{220}Rn exhalation rates were in the range of 3.73 - 326 mBq.m^{-2}.s^{-1} and 144 -9470 mBq.m^{-2}.s^{-1}, respectively. The arithmetic average value of the ^{222}Rn and ^{220}Rn

Table 1. Radon and Thoron Exhalation Rates from Surface Soil in Bangka-Belitung Islands

Site	GPS		Radon-Thoron Exhalation Rate ($Bq\ m^{-2}s^{-1}$)		Parent Nuclide Concentrations (Bq/kg)	
	S	E	^{222}Rn	^{220}Rn	^{226}Ra	^{232}Th
1.	2.05341	105.96299	97.94 ± 19.04	177 ± 26	39.0 ± 2.6	75.7 ± 2.1
2.	1.92487	105.73072	3.73 ± 0.58	194 ± 29	26.2 ± 2.0	21.3 ± 1.6
3.	1.86887	105.55734	20.00 ± 3.54	196 ± 29	16.8 ± 1.3	28.0 ± 0.9
4.	1.90731	105.38345	42.93 ± 7.23	927 ± 137	116.3 ± 7.2	219.7 ± 5.9
5.	2.05292	105.17896	24.08 ± 3.46	947 ± 140	136.4 ± 8.5	601.2 ± 33.3
6.	2.02674	106.11205	17.98 ± 3.78	303 ± 45	29.5 ± 2.1	62.5 ± 1.8
7.	1.83548	106.09581	33.84 ± 4.78	4659 ± 691	143.7 ± 9.0	377.4 ± 10.2
8.	1.74349	105.93686	13.37 ± 2.85	2696 ± 400	80.6 ± 5.1	252.6 ± 6.8
9.	1.65345	105.80455	18.52 ± 3.34	3902 ± 579	76.7 ± 5.1	231.0 ± 6.6
10.	2.42239	106.30708	5.63 ± 1.31	1106 ± 210	63.0 ± 4.1	151.7 ± 4.2
11.	2.48396	106.41884	34.95 ± 6.06	162 ± 24	23.0 ± 1.6	44.3 ± 1.3
12.	2.61175	106.36892	291 ± 53	9470 ± 1404	543.8 ± 36.3	2170 ± 65.2
13.	2.79730	106.41721	61.03 ± 9.05	1644 ± 244	118.4 ± 7.4	510.8 ± 13.6
14.	3.00411	106.47252	20.27 ± 1.86	1969 ± 419	91.2 ± 5.9	109.0 ± 6.4
15.	2.99088	106.60519	48.21 ± 6.33	1875 ± 529	54.5 ± 3.6	115.0 ± 0.5
16.	2.70761	106.30343	7.96 ± 1.29	895 ± 168	64.4 ± 4.3	155.0 ± 4.3
17.	2.72320	106.17023	14.12 ± 1.60	564 ± 84	43.2 ± 3.8	77.8 ± 5.2
18.	2.61162	106.15139	46.43 ± 5.91	855 ± 127	46.0 ± 3.1	97.4 ± 2.7
19.	2.55059	106.48337	20.42± 1.97	3539 ± 864	42.1 ± 2.9	123.6 ± 3.4
20.	2.55455	106.64125	58.76 ± 10.23	1648 ± 487	99.6 ± 6.9	158.9 ± 4.6
21.	2.04609	105.76947	7.58 ± 1.52	3077 ± 813	115.2 ± 7.3	206 .8 ± 11.6
22.	1.99569	105.65130	19.32 ± 1.85	144 ± 21	25.4 ± 1.9	81.4 ± 2.3
23.	1.72616	105.45825	8.00 ± 1.47	164 ± 24	29.2 ± 2.0	59.0 ± 1.7
24.	1.64023	105.51582	30.46 ± 5.18	194 ± 29	22.9 ± 1.7	33.2 ± 1.0
25.	1.59386	105.57115	18.48 ± 3.14	3181 ± 825	61.6 ± 4.1	230.6 ± 13.8
26.	2.41710	106.05264	84.14 ± 17.72	5848 ± 1102	139.6 ± 8.5	412.7 ± 10.7
27.	2.76905	107.71902	22.15 ± 2.91	214 ± 32	71.8 ± 4.7	90.9 ± 2.6
28.	2.80287	107.76521	22.22 ± 3.03	2527 ± 617	68.3 ± 4.5	212.0 ± 6.0
29.	2.92091	107.82620	111 ± 11	4306 ± 947	194.8 ± 12.2	492.8 ± 28.7
30.	3.02891	107.89948	4.40 ± 0.63	254 ± 32	10.7 ± 0.9	18.4 ± 0.6
31.	3.09563	107.99509	87.18 ± 8.86	1820 ± 270	178.2 ± 11.0	376.7 ± 10.2
32.	2.70687	107.65446	18.07 ± 3.60	183 ± 27	57.6 ± 3.7	94.0 ± 2.6
33.	2.92137	108.18832	35.78 ± 3.43	1353 ± 256	89.5 ± 5.7	188.2 ± 5.1
34.	2.70462	108.02458	43.93 ± 7.44	3708 ± 550	101.5 ± 6.3	328.4 ± 19.1
35.	3.19425	107.59635	12.12 ± 1.43	199 ± 53	46.8 ± 3.1	99.6 ± 2.8
36.	2.75909	107.82616	326 ± 19	7398 ± 1097	258.1 ± 15.7	653.8 ± 38.0
Range			3.73 – 326	144 – 9470	10.7 – 543.8	18.4 - 2170
Average			48.11	2008	92.38	254.5

exhalation rates by all thirty six data were obtained to be 48.11 $mBq.m^{-2}.\ s^{-1}$, 2008 $mBq.m^{-2}.\ s^{-1}$, respectively. The distribution of ^{222}Rn and ^{220}Rn exhalation rates was indicated in Figure 4. This figure shows that ^{222}Rn and ^{220}Rn exhalation rates vary widely from site to site.

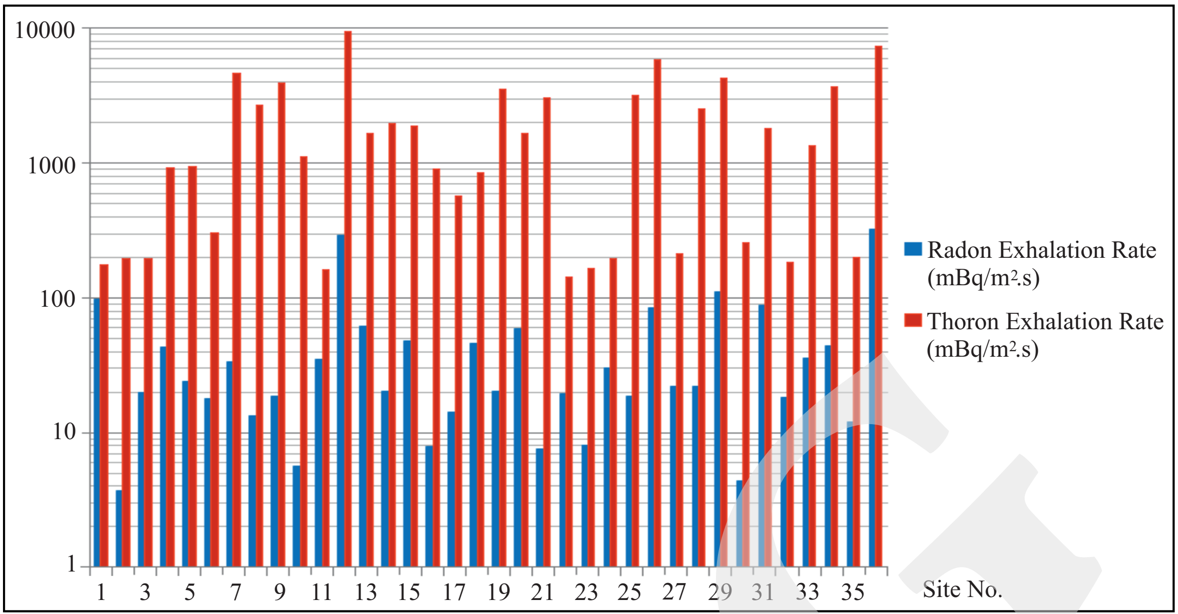

Figure 4. The distribution graph of radon and thoron exhalation rate levels.

Seventy five percent of ^{222}Rn exhalation rate measurement results are more than 10 mBq.m^{-2}.s^{-1} and 8.3 % of its are more than 100 mBq.m^{-2}.s^{-1}, however 7.5 % of its are lower than 10 mBq.m^{-2}. s^{-1}. Most of ^{220}Rn exhalation rates are higher than ^{222}Rn exhalation rate, where 47.2 % of its are more than 100 mBq.m^{-2}.s^{-1} and 52.8 % of results had a exhalation rate of more than 1000 mBq.m^{-2}. s^{-1}. It was admitted that the ^{220}Rn exhalation rate was about forty two times of the ^{222}Rn exhalation rate.

In order to evaluate how ^{222}Rn and ^{220}Rn exhalation rates are influenced by the activity concentration of their precursors (^{226}Ra and ^{232}Th, respectively), the variation of ^{222}Rn and ^{220}Rn exhalation rates were observed to variations of radium and thorium concentrations in soil collected from the same site. The concentration of ^{226}Ra and ^{232}Th in soil samples collected from the same measurement sites were shown in Table 1, columns 6 and 7. Generally, the distribution of ^{226}Ra and ^{232}Th in soil showed the same tendency as ^{222}Rn and ^{220}Rn distribution. By using its concentration levels, the correlation between the ^{222}Rn exhalation rate and the ^{226}Ra concentration and between the ^{220}Rn exhalation rate and the ^{232}Th concentrations were presented as in Figures 5 and 6.

As can be seen in Figures 5 and 6, it was found a weak correlation between the ^{222}Rn

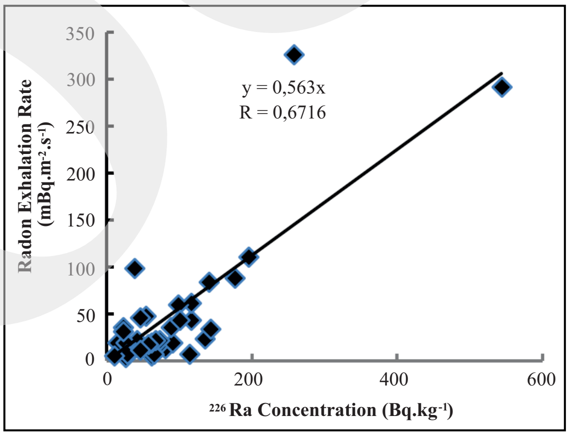

Figure 5. Correlation between ^{222}Rn exhalation rate and parent ^{226}Ra concentration.

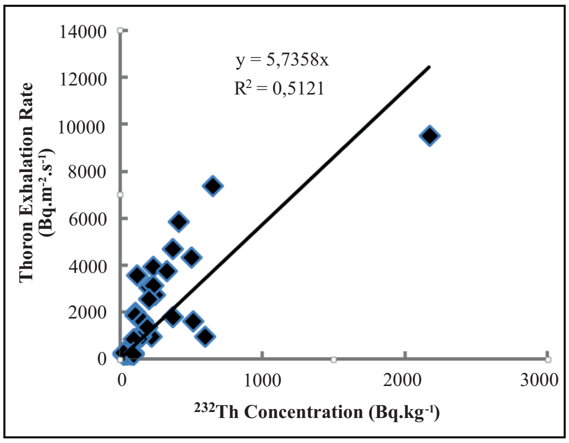

Figure 6. Correlation between ^{220}Rn exhalation rate and parent ^{232}Th concentration.

exhalation rate and the radium concentration (R = 0.67), and between the ^{220}Rn exhalation rate and the thorium concentration (R = 0.51). The weak correlation may be influenced by environmental factors, such as weather, water content of soil, and geology. According to Sun *et al.* (2004), the main factors influencing ^{222}Rn diffusion in soil are the soil characteristics such as soil porosity and moisture. Advection takes place when there is pressure difference between the airs of pore space and ground surface. The most important factor affecting advection is the soil permeability. Other meteorological parameters like temperature difference between soil and surface air, wind velocity, and rainfall also affect the advection process. Hosoda *et al.* (2007) reported that the exhalation rate showed a decreasing tendency for the increase in the moisture content over 8 %.

At the measurement day, the soil condition was different from each other such as dry, semi-moist, and wet. The weather conditions on the day of measurement were partly cloudy, partly sunny, and cloudy. Based on the conducted studies, it was observed that with reduced soil moisture and weather conditions changing into sunny, ^{222}Rn and ^{220}Rn exhalation rates increase.

In comparison to a world average, the results indicate that the ^{222}Rn and ^{220}Rn exhalation rates for the studied area are much higher than the worldwide average. UNSCEAR (2000) report shows that the world averages of the ^{222}Rn and ^{220}Rn exhalation rates are 26.2 mBq.m^{-2}. s^{-1} and 1000 mBq.m^{-2}.s^{-1}, respectively. Thus, the obtained value in this study were twice of values shown in UNSCEAR (2000) report. This fact suggests that the ^{222}Rn and ^{220}Rn exhalation rates in Bangka-Belitung Islands (Figure 7) must be considered to assess the radiological hazard of living in these areas.

^{222}Rn and/or ^{220}Rn exhaled from the surface soil may migrate into the structure of dwelling and accumulate indoors in sufficient quantities to pose a health hazard. WHO (2009) has classified them as carcinogenic to humans and the second most important cause of lung cancer after cigarette smoking. WHO (2009) recommended the levels of radon in the residental buildings as 100 Bqm^{-3}.

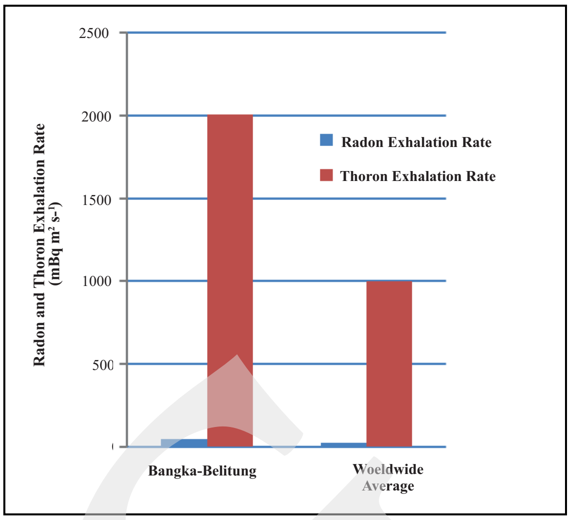

Figure 7. Comparison of ^{222}Rn and ^{220}Rn exhalation rates in Bangka-Belitung with worldwide average values.

CONCLUSSION

The ^{222}Rn and ^{220}Rn exhalation rates from surface soil of Bangka and Belitung Islands have been determined in situ measurement by using an accumulation chamber equipped with a solid-state alpha particle detector of RAD7. Then, the activity concentrations of parent radionuclides (^{226}Ra and ^{232}Th) in soil samples collected from the same site have been determined in the laboratory by using gamma-ray spectroscopy. The result of measurement showed that ^{222}Rn and ^{220}Rn exhalation rates as well as activity concentrations of ^{226}Ra and ^{232}Th varied widely from site to site. Mostly, the distribution of ^{226}Ra and ^{232}Th showed the same tendency as ^{222}Rn and ^{220}Rn distribution, but it was not any strong correlation due to the influence of environmental factors, such as weather, water content of soil, pressure, temperature, and humidity conditions. All the measurement result showed that the ^{220}Rn exhalation rate was higher than the ^{222}Rn exhalation rate. From this study, it was also found that Bangka Belitung Islands have the ^{222}Rn and ^{220}Rn exhalation rate higher than the world average value reported by UNSCEAR. The geology of Bangka Belitung Islands is covered by granite which has a higher radioactivity concentration level than the soil from the common areas world average. Due to high level of ^{222}Rn and ^{220}Rn exhalation rates in some areas of the

BangkaBelitung Islands, necessary provisions should be considered for construction in these areas to avoid ^{222}Rn and ^{220}Rn entrance into residential buildings.

Acknowledgment

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