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^{222}Rn Emanation and Exhalation Rate of Soil from Tasik Paya Bungor Area using CR-39 and Continuous Radon Monitor

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Of late, environmental pollution caused by outdoor ^{222}Rn emanation rate from soil has become one of the main concerns of many researchers. This research focuses on the ^{222}Rn emanation rate of Naturally Occurring Radioactive Materials (NORM) in the soil from the Tasik Paya Bungor area, Malaysia, which is one of the oldest tropical rainforest in the world. This area has been transformed into a plantation area by the locals. With the increment of agricultural activities in the area which involve the use of phosphate fertilisers and pesticides, the radiation contamination due to ^{222}Rn emanation rate from the soil has also increased. To investigate the ^{222}Rn emanation rate from the soil, samples from 14 different points on selected locations were collected using a hand auger, based on a standard sampling method. Both methods in determining the ^{222}Rn emanation rate for long-term measurement using CR-39 and ^{222}Rn exhalation rate using diurnal measurement by Sun Nuclear Continuous Radon Monitor, CRM-1029 in Air-Tight Chamber were used. From the results, the average ^{222}Rn emanation rate obtained using CR-39 is 0.51±0.05 Bq m^{-3} day^{-1}. As for the diurnal measurement using CRM-1029, the ^{222}Rn mass exhalation rate reading ranges between 0.11 – 0.72 mBq kg^{-1} hr^{-1} and the ^{222}Rn surface exhalation rate between 0.31 - 2.0 Bq m^{-2} hr^{-1}. It shows that both methods may be used, depending on the accessibility. The values are considered low as compared to the allowed national limit. However, in the future, more points on the selected locations should be considered for radon emanation rate measurement and its natural radionuclides concentration should also be determined.

Keywords: CR-39; CRM-1029; NORM; Radon emanation rate; SSNTD

I. INTRODUCTION

The inhalation of radon gas and its progeny is known as one of the main routes of radiation exposure to human population (UNSCEAR, 2000). This colourless, odourless, and radioactive chemical element exists naturally in the earth's crust (Cevik *et. al.*, 2011; Gilmore *et al.*, 2002). Thorium and uranium undergo radioactive decay to produce stable lead and in between the process, release radon gas. Having a half-life of 3.8 days, ^{222}Rn is produced from the decay of ^{226}Ra, through ^{238}U decay series and diffuses through soil into the atmosphere (Singh *et al.*, 2004). Even though ^{222}Rn has a very short half-life, its presence in the environment has been common due to its continual regeneration. The main contributors to its regeneration are thorium and uranium, which are abundant on Earth and have long half-lives. As an inert gas, ^{222}Rn releases electrically charged particles from its short half-lived progenies. Hence, it can be attached to natural aerosol and dust which if inhaled, will tend to spread into the lungs. This may lead to health risk as it may expose the lung epithelial bronchial cells to excessive alpha radiation (Al-Zoughool *et al.*, 2009).

In this study, soil samples were collected from the area of Tasik Paya Bungor area whereat many research focusing

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mainly on fisheries and aquaculture, water quality analysis and flora and fauna have been conducted (Sharip *et al.*, 2007). Due to the increment of agricultural activities in the area, outdoor radon investigations need to be conducted to study the ^{222}Rn emanation and exhalation rates from the soil within the area. ^{222}Rn emanation happens due to the formation of radon atoms through radium decay from the grain which are then released into the interstitial spaces between the grains while ^{222}Rn exhalation occurs due to the transportation of radon atom to the ground surface before being exhaled to the atmosphere (Ishimori *et al.*, 2013). Figure 1 summarises the difference between radon emanation and exhalation activities within the soil.

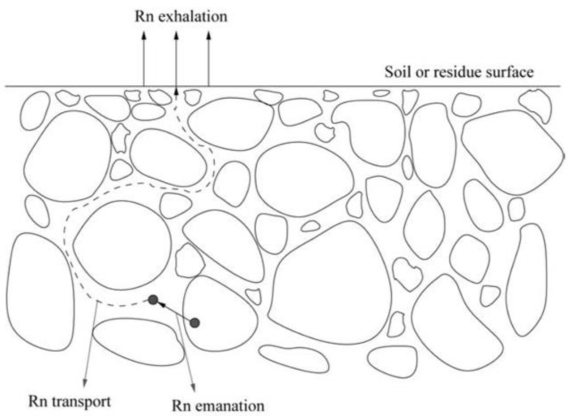

Figure 1. Processes leading to radon release to the atmosphere (Ishimori *et al.*, 2013)

The soil samples collected were prepared for ^{222}Rn emanation rate study using CR-39 and ^{222}Rn exhalation rate study using Sun Nuclear Continuous Radon Monitor, CRM-1029. This study focuses on the ^{222}Rn emanation and exhalation rates from the soil in an air-tight chamber.

II. MATERIALS AND METHOD

The study was conducted in the Tasik Paya Bungor area. 14 points near Tasik Paya Bungor, Gambang were selected based on the grid method. The soil at each point was dig 10 cm under the ground level using a hand auger and the detectors were placed inside the points. Table 1 shows the coordinates of the samples at the selected points obtained by using a global positioning system (GPS).

Table 1. Coordinates of soil samples at the selected points in Tasik Paya Bungor area

Point	Latitude, N	Longitude, E	Location Description
1	3° 42' 57.65"	102° 55' 49.71"	Near railroad
2	3° 42' 58.00"	102° 55' 52.41"	Near railroad
3	3° 42' 51.00"	102° 55' 59.90"	Inside jungle
4	3° 42' 49.23"	102° 55' 57.78"	Inside jungle
5	3° 42' 49.23"	102° 56' 00.49"	Inside jungle
6	3° 42' 49.23"	102° 56' 03.19"	Palm tree area
7	3° 42' 49.23"	102° 55' 05.93"	Palm tree area
8	3° 42' 46.43"	102° 55' 55.12"	Inside jungle
9	3° 42' 46.43"	102° 55' 57.84"	Inside jungle
10	3° 42' 46.43"	102° 56' 03.80"	Inside jungle
11	3° 42' 46.43"	102° 56' 05.88"	Palm tree area
12	3° 42' 43.64"	102° 55' 57.82"	Inside jungle
13	3° 42' 43.64"	102° 56' 00.50"	Inside jungle
14	3° 42' 43.64"	102° 56' 03.16"	Inside jungle

CR-39 thin film, a solid-state nuclear track detector by Tastrak UK was used to detect the alpha radiation of ^{222}Rn. Its polymer surface structure would collide with the alpha particle and create trails of chemical bonds on the structure surface. The trails were enhanced after being chemically etched. From these trails, the ^{222}Rn track density may be observed and by calculation, the ^{222}Rn concentration from the soil samples may be obtained, hence the emanation rate may be determined.

The CR-39 thin films were designed into 1 cm x 1 cm dimension before being placed in radon monitors adopting can-mode configuration (Sani *et al.*, 2018) as shown in Figure 2. Sealed-cup radon monitors were prepared by following Sani *et al.* (2018).

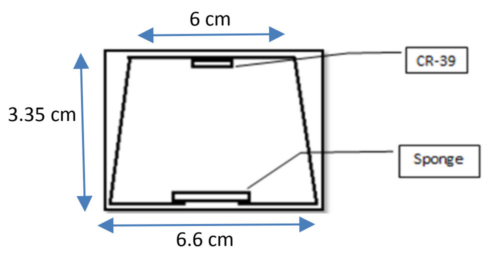

Figure 2. Placement of CR-39 detector inside a sealed-cup radon monitor

The CR-39 detector in radon monitors were exposed to the soil at roughly 8.35 cm below the ground level for 30 days to investigate the ^{222}Rn emanation rate emitted from the soil at each point. Figure 3 shows the setup of each CR-39 detector when exposed to the soil at every selected point.

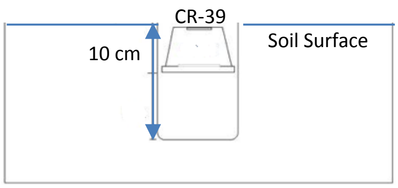

Figure 3. Schematic diagram of the placement of radon monitor in soil at each point

After 30 days, the radon monitors were retrieved and CR-39 were etched with 6 M NaOH at 70 °C for 6 hours. A minimum of one-hour rinse with running cold water was required to make the ^{222}Rn tracks visible on the surface of the detector (Sani *et al.*, 2018). After etching, CR-39 was observed under Leica Optical Microscope at 10X magnification and the ^{222}Rn track density calculation was conducted from the ^{222}Rn emanation rate. The observed sections of diameter 50 μm were taken for calculation. Figure 4 shows the dark spots observed.

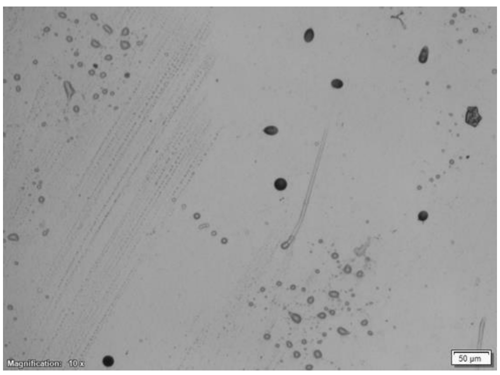

Figure 4. Optical image of dark spots observed on CR-39 detector

The calculation of ^{222}Rn track density, ρ [track / m^2] visible on CR-39 under optical microscope and its track density frequency, X [track / m^2.day] were determined by Equation (1) and (2) (Al-Kaabi *et al.*, 2015):

$$\rho = \frac{Average\ total\ tracks}{Area\ of\ field\ view} \tag{1}$$

$$X = \frac{\rho}{days} \tag{2}$$

From ^{222}Rn track density frequency, the ^{222}Rn concentration, C_a [Bq / m^3] and emanation rate, E_m [Bq / m^2.day] were determined using Equation (3) and (4), respectively (Al-Kaabi *et al.*, 2015):

$$C_a = X\ x\ F \tag{3}$$

$$E_m = \frac{C_a\ x\ V}{A\ x\ days} \tag{4}$$

where F is the standard soil constant per track, 7.152 x 10^{-2} Bq track^{-1} day^{-1}, V is air in cup volume in m^3 and A is the hole area of the opening in m^2. The standard soil constant, F has been determined earlier during calibration.

The ^{222}Rn concentration was measured using the specifically patented Sun Nuclear Continuous Radon Monitor-1029. Its high sensitivity makes it suitable for ^{222}Rn indoor measurement. It also measures the temperature, humidity and pressure of measured environment. For this experiment, CRM-1029 was set at the interval of every one hour for 72 hours of exposure. In order to avoid any effect of electrostatic charges, the sample container used for this study was made of plastic. The volume of the container is 0.04073 m^3, following the dimension conducted by previous study methods (Khalid *et al.*, 2014). Leakage of gas from the inside of the container to the outside or vice versa may be avoided by ensuring that the container was sealed tightly using masking tape every time the experiment was conducted. For every change of sample, the background radiation, C_b was measured. The measurements were conducted three times over three days for every sample to ensure its accuracy. Figure 5 shows the schematic diagram of the experimental setup.

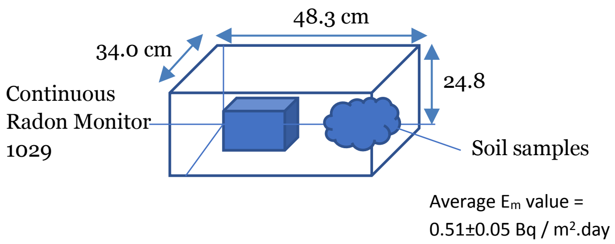

Figure 5. Schematic diagram of the radon air-tight chamber

Environmental Protection Agency (EPA) average reading was taken directly from the data produced by the CRM-1029 monitor, showing the average radon emanation for 72 hours of measurement at every hour interval. To ensure data accuracy, the results obtained were then subtracted to the background reading, C_b. The corrected ^{222}Rn activity concentration, C_{Rn} and ^{222}Rn concentration in secular equilibrium, C_{eq} were obtained by using Equation (5) and (6), respectively (Hussein *et al.*, 2012):

$$C_{Rn} = C_{Ave} - C_b \quad (5)$$

$$C_{eq} = \frac{C_{Rn}}{(1-e^{-\lambda t})} \quad (6)$$

The results were then tabulated, analysed and the ^{222}Rn mass and surface exhalation rate, E_m [Bq/ kg.hr] and E_S [Bq / m².hr] were calculated using Equation (7) and (8), respectively (Hussein *et al.*, 2012):

$$E_m = \left(\frac{C_{eq}\lambda_{Rn}V_{eff}}{m}\right) \quad (7)$$

$$E_S = \left(\frac{C_{eq}\lambda_{Rn}V_{eff}}{S}\right) \quad (8)$$

where λ_{Rn} is ^{222}Rn decay constant, 7.554 x 10^{-3} hr^{-1}, t is the time of the ^{222}Rn activity build-up in the container in hr, V_{eff} is air volume inside the air-tight container in m³, m is mass of the sample in kg and S is surface area of sand brick in m².

III. RESULTS AND DISCUSSION

The results in location 6, which is a palm tree area, showed the highest ^{222}Rn concentration hence the biggest contributor to ^{222}Rn emanation rate, 1.17±0.16 Bq / m².day followed by the results in location 10, which is inside a jungle, with ^{222}Rn emanation rate 1.11 ± 0.036 Bq / m².day. The lowest ^{222}Rn concentration soil was from location 3 and the soil sample from the jungle area, with ^{222}Rn emanation rate 0.14±0.01 Bq / m².day. Figure 6 shows the ^{222}Rn emanation rate of soil samples from all points selected. It shows that soil samples taken from a commercial agricultural area contributes more ^{222}Rn emanation rate to the surrounding which could be a result of the usage of phosphate fertilisers.

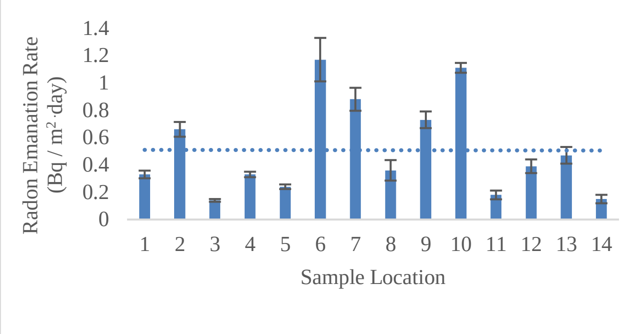

Figure 6. ^{222}Rn emanation rate of soil samples from selected locations

Table 2 tabulates the comparison between ^{222}Rn emanation rate from the study to others conducted around the world. The values obtained were lower compared to other studies from different places as well as the World Limit of 3.43 Bq / m².day (ICRP, 1993). Many factors contribute to ^{222}Rn concentration and emanation coefficient such as macroscopic properties, grain permeability, differences in radium concentrations, radium distribution, grain surfaces crystallisation, as well as the grain texture and size (Ahmad *et al.*, 2014). One of the most important factors that influence the radon concentration is the radium distribution within the grains. The measured ^{222}Rn concentration will be automatically increased if the radium distribution concentrated on the thin grain surface layers. This leads to the increment of ^{222}Rn atoms diffusion into the pore spaces. Even though the amount of radium concentration in the particles are similar, its emanation rates are higher in frangible rather than in solid grains (Hassan *et al.*, 2016).

Table 2. Comparison of ^{222}Rn emanation rate from soil in different locations

Location	**Average ^{222}Rn Emanation Rate (Bq m^{-2} day^{-1})**	**Reference**
Tasik Paya Bungor, Gambang	0.51±0.05	Present study
Bahawalpul Division, Pakistan	1.56 – 3.33	Rahman, S., 2006
NWFP, Pakistan	2.49 – 4.66	
Turkey	0.4	Baykara *et al.*, 2005
Azad Kashmir, Pakistan	0.17 – 0.34	Rafique *et al.*, 2011

Figure 7 shows the comparison between ^{222}Rn mass and surface exhalation rate from the same samples at selected locations. The values were calculated from ^{222}Rn concentration measured using CRM-1029 for samples from each location studied. It shows that the ^{222}Rn mass exhalation rate reading averages between 0.11 – 0.72 mBq kg^{-1} hr^{-1} and the ^{222}Rn surface exhalation rate between 0.31 - 2.0 Bq m^{-2} hr^{-1}.

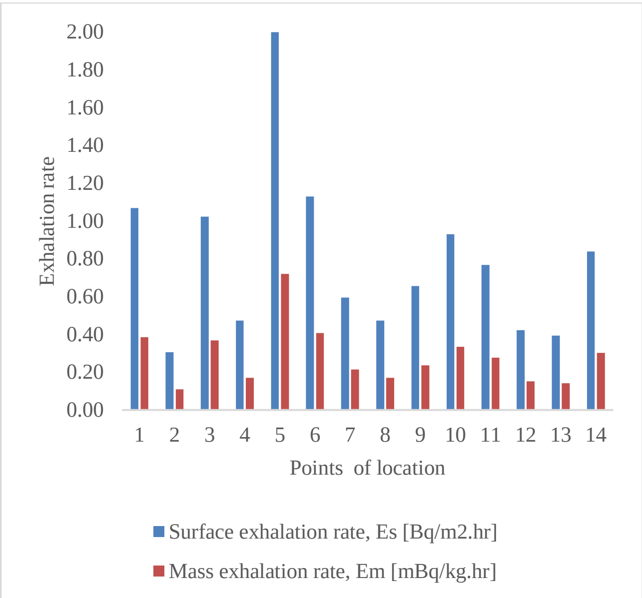

Figure 7. ^{222}Rn Mass Exhalation Rate (E_m) and Surface Exhalation Rate (E_s) from soil samples at selected points of research

Table 3 compares the average ^{222}Rn mass and surface exhalation rate from the study to other values around the world. It shows that both mass and surface exhalation rate from the soil of Tasik Paya Bungor are still within the safe limit and averages with the results from other countries. The values are also within the World Limit of 37.8 mBq kg^{-1} hr^{-1} (UNSCEAR, 1988; Chowdhury *et al.*, 1999)

Table 3. Comparison of ^{222}Rn mass and surface exhalation rate from soil in different locations

Location	**Average ^{222}Rn Mass Exhalation Rate, E$_m$ (mBq kg^{-1} h^{-1})**	**Average ^{222}Rn Surface Exhalation Rate, E$_s$ (Bq m^{-2} h^{-1})**	**Reference**
Tasik Paya Bungor, Gambang	0.11 – 0.72	0.31 - 2.0	Present study
Benghazi, Libya	8.2±0.3	0.22±0.01	Saad *et al.*, (2013)
Al-Marj, Libya	13.9±0.5	0.32±0.01	
Southern Egypt	NA	14.12 - 59.44	Sroor *et al.*, (2001)
USA	7.6	NA	Nazaroff *et al.*, 1992
Himalaya, India	0.91±0.16	NA	Prasad *et al.*, 2008
Chittagong, Bangladesh	1.06±0.59	1.58±0.89	Chowdhury *et al.*, 1999
Northern Peninsular Malaysia	0.19 – 0.88	NA	Almayahi *et al.*, 2013
World Limit	37.8	0.96	UNSCEAR, 1988; Chowdhury *et al.*, 1999

IV. CONCLUSION

This paper has presented the ^{222}Rn emanation and exhalation rates of soil to the environment in Tasik Paya Bungor area. In general, the ^{222}Rn emanation rate obtained was lower compared to the global average values. With the world ^{222}Rn emanation rate limit of 3.43 Bq m^{-2} day^{-1}, the highest value determined in this study is still much lower. It has been proven that the natural limit was not reached for the study area and it shows no significant difference from other regions of the world. Hence, for natural radioactivity mapping, the result may be considered a useful resource to be referred to. More samples should be taken from other locations around the Tasik Paya Bungor area and from all over Malaysia especially with soils from other lake-type locations. For further investigations, besides using soil as sample, researchers may investigate other elements such as plants, sediments and water from the place of study to determine the ^{222}Rn concentration from it. From the results, radiation safety level may be determined and further actions by the authorities may be taken accordingly. The radiological quality of the soil around Tasik Paya Bungor area may be obtained from this study and the authorities can create radiation related awareness to the public within this area. To avoid any excessive radiation from direct exposure, future construction developers may take necessary precautions before further development of this area and take mitigation actions at locations with high radioactivity concentration. Moreover, the findings may be used by the authorities to design rules and laws for the incoming plantation and agricultural activities and for future researchers who will embark in environmental radiation field.

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