Risk assessment of ^{222}Rn in water resources in Malacca, Malaysia: Consequences for public consumption

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Abstract. The primary motivation for this work is to evaluate the ^{222}Rn concentration in the various water resources at Malacca, Malaysia. In this study, the measurement was carried out using a portable DURRIDGEs' RAD7 detector. The measured ^{222}Rn concentration was found to be in the range of (0.07 ± 0.12 to 111.35 ± 2.76) Bq/1**,** with an average is 21.34 ± 1.51 Bq/l. From this data, the age-dependent associated Annual Effective Dose (AED) due to the ingestion and inhalation of ^{222}Rn from the consumption of drinking water were calculated for the evaluation of the public health risk from the ^{222}Rn exposure. The total AED due to ingestion and inhalation of water were to range from (0.001 to 0.87) mSv y^{-1} with an average value of 0.167 mSv y^{-1} for infant, (0.0003 to 0.497) mSv y^{-1} with an average value of 0.095 mSv y^{-1} for children and (0.0004 to 0.565) mSv y^{-1} with an average value of 0.108 mSv y^{-1} for adult. The total AED for group of infant and adult in this study were exceed the limit recommended by WHO and IAEA, but the value is significantly lower than the limit ICRP. However, the obtained data could serve as a reference for any future radiological study especially for ^{222}Rn concentration in water resources at Malacca, Malaysia.

Keywords: RAD7, Water, ^{222}Rn concentration, Annual Effective Dose (AED)

INTRODUCTION

Water is one of the natural resources that very important for most life on this earth. In water sources, it usually contains many naturally occurring radionuclides such as Radon-222 (^{222}Rn), Radium-226 (^{226}Ra), Uranium-238 (^{238}U), Thorium-232 (^{232}Th) and Actinium-228 (^{228}Ac) isotopes, etc. All the concentration of radionuclides is highly variable as they depend on the nature of the aquifer and the prevailing lithology around it (Rangaswamy *et al.* 2016).his is the standard font and layout for the individual paragraphs.

^{222}Rn (half-life, $T_{1/2}$ = 3.82 days), is a ubiquitous, colorless, tasteless and odorless is derived from the decay of ^{238}U series radionuclides. About 99.3% of all uranium in the crust; it is a rich and dangerous radioactive isotope in the natural environment (Rangaswamy *et al.* 2016).

Besides that, the ^{222}Rn can be found in everywhere in the air, water, rocks and soils of the Earth's crust. The occurrence of natural radionuclides especially ^{222}Rn in drinking water causes threats to human health such as lung cancer, colon cancer and leukemia when it taken into body due to ingestion and inhalation (*Rangaswamy et al.* 2016). Through the epidemiological studies, it have shown some evidence of stomach cancer when the intakes of ^{222}Rn via the ingestion (P. Ravikumar and R.K. Somashekar, 2014; H. Nuhu *et al.* 2020). According to EPA, ^{222}Rn is the second leading cause of lung cancer and smoking being the first. USEPA has proposed the value of 11.1 Bq/l as the allowed maximum contamination level (MCL) for ^{222}Rn concentration in water (USEPA, 1999). The World Health Organization (WHO) also setup up the reference limit for safe drinking water is 100 Bq/l (WHO, 2006). In addition, the reference dose for intakes of ^{222}Rn in drinking water also has been setup by certain organization. The International Commission on Radiological Protection (ICRP) has recommended the reference level corresponding to an annual effective dose (AED) in the range of 1 to 20mSv with the consideration value of the order of 10 mSv y^{-1} should be the benchmark for setting a reference level for ^{222}Rn exposure (ICRP, 2015; H. Nuhu *et al.* 2020). The WHO and International Atomic Energy Agency (IAEA) were also recommended the reference limit for AED is 0.1 mSv y^{-1} (WHO, 2006; D. S. Guide, 2002).

In worldwide, the monitoring of ^{222}Rn content in drinking water has been increased for the radiation protection purpose. Furthermore, there are several research of ^{222}Rn concentration in drinking water have been conducted in some part of the world such in Indonesia (E.D. Nugraha *et al*. 2021), India (Rangaswamy *et al.* 2016), Yemen (A.I.A. El-Mageed *et al*, 2018), Saudi Arabia (E.H. El-Araby *et al*. 2019), Iraq (A.A. Ibrahim *et al.* 2017), etc. However, the research related to ^{222}Rn dose exposure among different age groups consist of infant, children and adult is still limited. Hence, this research presents the ^{222}Rn concentration in 17 samples of water collected from various water resources in Malacca, Malaysia. Further, dose assessments to International Commission on Radiological Protection (ICRP) proposed age groups of infant (1-2 years), children (2-17 years) and adult (> 17 years) were calculated based on the result obtained from the measurement of ^{222}Rn concentration.The style is called "Paragraph."

STUDY AREA

This study was conducted in years 2019 at Malacca, Malaysia. The total land area in Malacca is about 1664 km^2 and the area was covered with three districts of Jasin, Melaka Tengah and Alor Gajah. It has a total population of 932,700 in year 2020. A total of 17 samples of water samples were collected from several types of water resources such as lake water, sea water, hot spring water, river water, groundwater and tap water. The coordinate of sampling location was determined using a Global Positioning System (GPS) and the accurate coordinate for each sampling point was recorded for further reference. All the sampling information including the location, coordinates and types of water resources was provided in Table 1.

TABLE 1. Annual water consumption and dose conversion factor for different groups of age (UNSCEAR, 2000).

Sampling Code	Sampling location	Coordinate	Type of water resources
MC1	Taman Perdana Alai	2.172, 102.308	River

MC2	Kg. Ganun	2.409, 102.238	Hot spring
MC3	Bukit China	2.198, 102.256	Groundwater
MC4	Klebang	2.218, 102.188	Tap
MC5	Durian Tunggal	2.35, 102.25	Groundwater
MC6	Tanjung minyak	2.286, 102.197	Tap
MC7	Tanjung Bidara	2.293, 102.087	Sea water
MC8	Jasin	2.315, 102.431	Tap
MC9	Pulau Sebang	2.448, 102.236	River
MC10	Jasin	2.291, 102.377	Hot spring
MC11	Kampung Arongan	2.454, 102.237	River
MC12	Cherana putih	2.482, 102.179	Hot spring
MC13	Kg. Duyong	2.2, 102.297	Groundwater
MC14	Pangkalan Balak	2.321, 102.071	Sea water
MC15	Durian Tunggal	2.311, 102.32	Lake
MC16	Kampung Gangsa	2.285, 102.262	Tap
MC17	Taboh Naning	2.452, 102.181	Groundwater

Malacca State was selected as a study area because there is still has no study about ^{222}Rn concentration in the various water resources and this is a very interesting to explore. In Fig. 1, it shows the distribution of sampling areas in Malacca that collected from various water resources.

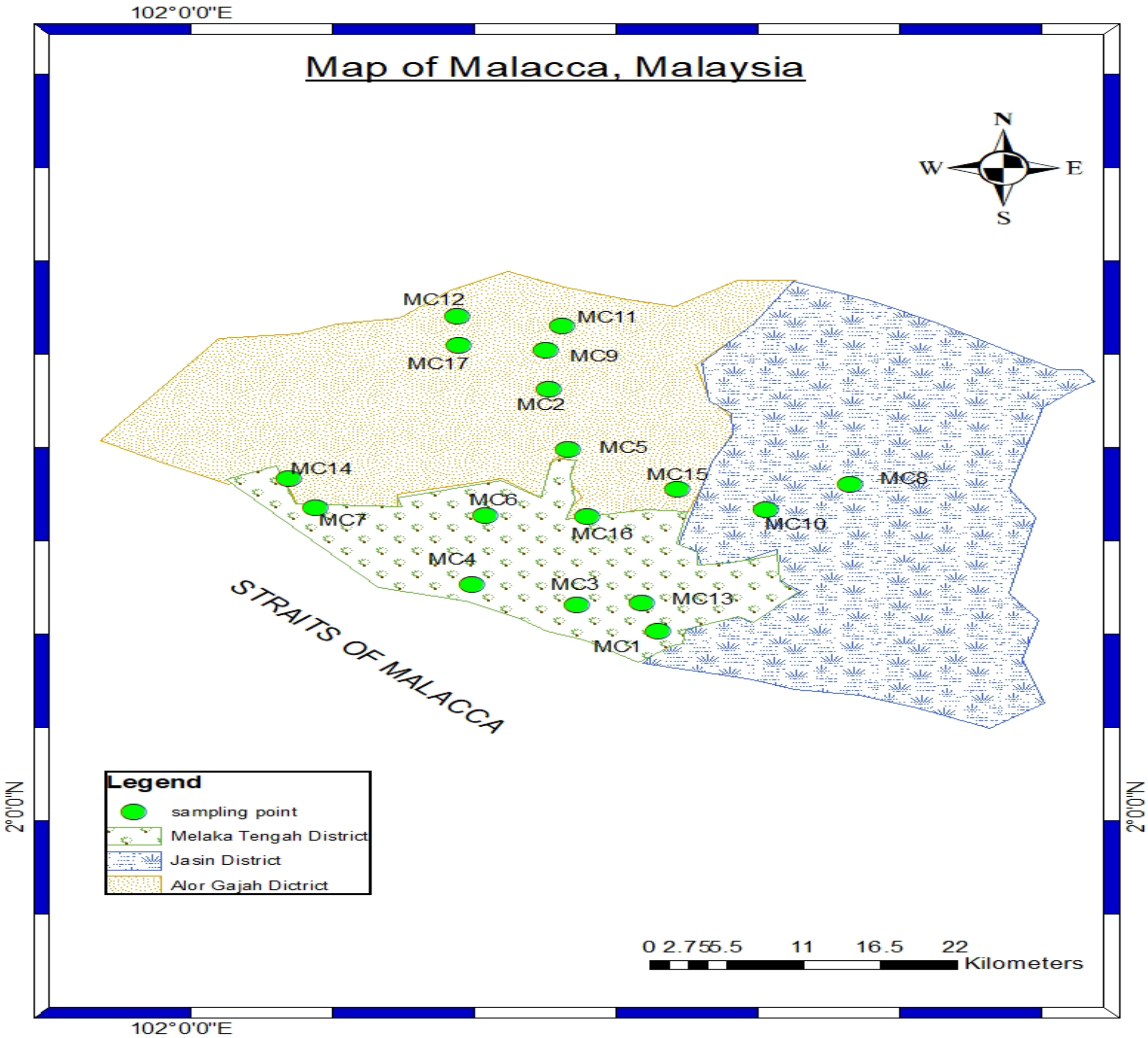

FIGURE 1. The distribution of sampling point in Malacca, Malaysia (using ArcGIS 10.4.1)

MATERIALS AND METHOD

Measurement of ^{222}Rn Concentration in Water

Seventeen (17) of water samples from various water resources were collected in the years of 2020. Sampling collected from private well or private area was done with permission from owners. The water samples were taken using well-washed vial of 250 ml and cap the vial while still under water in order to minimize the ^{222}Rn release to the air. Tighten the cap and the sample was labelled with the information such date, time, sampling location code and coordinates. The measurement of ^{222}Rn concentration in water was carried out using a calibrated portable ^{222}Rn monitor, RAD7.

In Fig. 2**,** it shows the schematic diagram setup in a closed air loop for the measurement of ^{222}Rn concentration in water that consists of three components which are the RAD7 detector together with the RAD7 H20 accessories that consist of the water vial, aerator and tube of desiccant (Durridge, 2011).

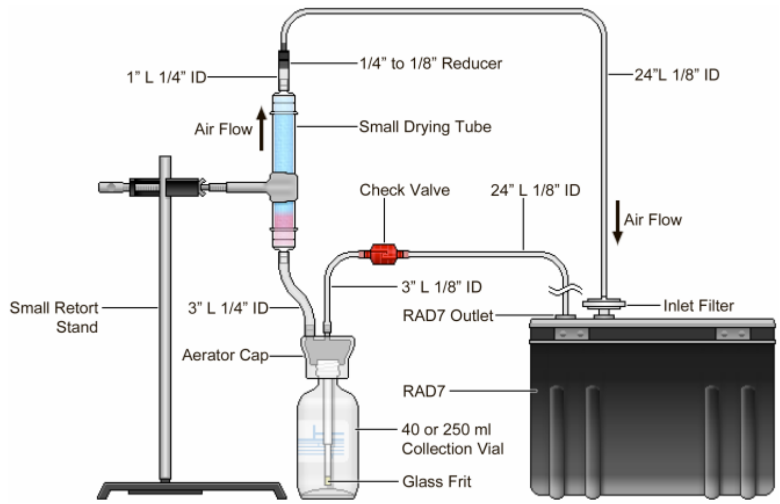

FIGURE 2. A schematic diagram of the RAD 7 and RAD7 H20 accessories (Durridge, 2011)

The internal pump with a flow rate of about 1 L min^{-1} will start run with five minutes of aerating the sample of water. At this aeration stage, the extraction efficiency or percentage of ^{222}Rn removed from the water to the air loop is very high with an approximately 94% until a state of equilibrium develops. Then, the ^{222}Rn gas will go through to the RAD7 detector after passing through the desiccant tube and air filter. The desiccant was used at all times to dry the air stream before it enters the RAD7. The RAD7 may give incorrect ^{222}Rn concentrations or may become damaged due to condensation on sensitive internal components if the desiccant is not used properly. The desiccant was very effective to maintain the relative humidity readings during the measurement remain at below 10%, as well as the detection efficiency of the RAD7 will decreases with the increasing of relative humidity (Durridge, 2011). The total time for complete measurement was about 30 minutes of counting and the summary of average ^{222}Rn activity in water was printed out by RAD7 infrared printer. In this measurement, the activity of ^{222}Rn was recorded in unit Bq m^{-3} (disintegration per second per m^3) or Bq/l (disintegration per second per litre) and the minimum detection limit of RAD7 for ^{222}Rn concentration in water is approximately 0.004 Bq/l (Durridge, 2015)

Assessment of Total AED From ^{222}Rn Exposure in Drinking Water

The total AED due to ingestion and inhalation of ^{222}Rn in drinking water for infants, children and adults are determined by the documentation of (UNSCEAR, 2000). The AED for ingestion, (AED$_{ing}$) was calculated according to equation (1):

$$AED_{ing}\ (mSv\ y^{-1}) = C_{Rn} \times A_i \times D_f \quad (1)$$

Where, C_{Rn} is the ^{222}Rn concentration in water (Bq/l), A_i is the annual water consumption (l y^{-1}). D_f is the ingesting dose conversion factor for ^{222}Rn (mSv Bq^{-1}). The values of C_{Rn}, A_i and D_f for groups of infant, children and adult set by (UNSCEAR, 2000) is listed in Table 2**.**

TABLE 2. Annual water consumption and dose conversion factor for different groups of age (UNSCEAR, 2000).

Age groups	Age range (y)	Annual water consumption, A_i ($l\ y^{-1}$)	Dose conversion factor, D_f ($mSv\ Bq^{-1}$)
Infant	1-2	150	2.3×10^{-5}
Children	2-17	350	5.9×10^{-6}
Adult	> 17	500	3.5×10^{-6}

When ^{222}Rn enters the human body, its short-lived progeny will decay, not only inside the stomach but also in the lungs. The AED to the lungs due to inhalation of ^{222}Rn in water was calculated using the parameters established in the UNSCEAR report (UNSCEAR, 2000). The AED for ingestion, (AED_{inh}) was calculated according to equation (2):

$$AED_{inh}\ (mSv\ y^{-1}) = C_{Rn} \times R_{AW} \times F \times O \times DCF \quad (2)$$

Where, C_{Rn} is the ^{222}Rn concentration in water (Bq/l), R_{AW} the ratio of ^{222}Rn in air to ^{222}Rn in tap water (10^{-4}), F the equilibrium factor for ^{222}Rn and its progeny (0.4), O the global average of indoor occupancy factor (7000 h y^{-1}) and DCF the dose conversion factor for ^{222}Rn exposure, 9 × 10^{-6} mSv h^{-1} Bq^{-1} m^3 (UNSCEAR, 2000).

RESULT AND DISCUSSION

The ^{222}Rn concentrations in various types of water resources are shown in Table 3. It is observed that the ^{222}Rn concentration ranges from (0.07 ± 0.12 to 111.35 ± 2.76) Bq/l**,** with an average for all sample is 21.34 ± 1.51 Bq/l. The minimum ^{222}Rn concentration was recorded in lake water that collected from the sampling location MC15 at Durian Tunggal and this value was extremely lower than the Maximum Contaminant Level (MCL) of 11.1 Bq/l. In this situation, the lake water was also recognized as a surface water and this expected to has a very low concentration of ^{222}Rn because of the ^{222}Rn gas contained in that surface water was easily escaped or released to the outdoor air (WHO, 2012). The maximum ^{222}Rn concentration was recorded in hot spring water that collected from the sampling location MC12 at Cherana Putih. This value was exceeded the MCL and World Health Organization (WHO) safe limit for drinking water. The MCL value proposed by United States Environmental Protection Agency (USEPA) is 11.1 Bq/l (USEPA, 1999) while the safe limit for drinking water is 100 Bq/l was recommended by WHO (WHO, 2006; H. Nuhu *et al.* 2020).

Furthermore, it is about 29.4 % of the samples that collected from the location of MC2, MC3, MC12, MC20 and MC17 were observed to have the ^{222}Rn concentration levels greater than 11.1 Bq/l. While, the ^{222}Rn concentration of others samples were recorded below this MCL limit. Compared to the proposed by WHO limit of 100 Bq/l for the safe guidance level of drinking water, all samples were below the acceptable limit, except for the sample MC12.

TABLE 3. The ^{222}Rn concentrations (Bq/l) in water from various type of water resources

Sampling location	Type of water resources	^{222}Rn concentration, C_{Rn} (Bq/l)

MC1 - Taman Perdana Alai	River	1.15 ± 0.38
MC2 - Kg. Ganun	Hot spring	88.33 ± 7.65
MC 3 - Bukit China	Groundwater	14.38 ± 1.84
MC4 – Klebang	Tap	3.78 ± 0.54
MC5 - Durian tunggal	Groundwater	7.42 ± 0.81
MC6 - Tanjung minyak	Tap	0.68 ± 0.31
MC7 - Tanjung Bidara	Sea water	1.38 ± 0.68
MC8 – Jasin	Tap	1.55 ± 0.36
MC9 - Pulau Sebang	River	4.30 ± 1.20
MC10 – Jasin	Hot spring	8.05 ± 0.92
MC11 - Kampung Arongan	River	9.63 ± 2.20
MC12 - Cherana putih	Hot spring	111.35 ± 2.76
MC13 - Kg. Duyong	Groundwater	18.52 ± 1.31
MC14 - Pangkalan Balak	Sea water	0.72 ± 0.52
MC15 - Durian Tunggal	Lake	0.07 ± 0.02
MC16 - Kampung Gangsa	Tap	2.30 ± 0.40
MC17 - Taboh Naning	Groundwater	89.12 ± 3.38
Average		21.34 ± 1.51

In Fig. 3, it shows the summarize graph of the average ^{222}Rn concentration in group of water resources and the reference line for the comparison. The average ^{222}Rn concentration hot spring water and groundwater obtained in the present study are 69.24 Bq/l and 32.36 Bq/l, respectively. Both ^{222}Rn concentrations in that water resource were compared to other resources from river water, tap water, sea water and lake. The comparison shows the ^{222}Rn concentration in hot spring and groundwater were much higher than other water resources and their concentration were also exceeded the reference limit of MCL as recommended by USEPA. In the meanwhile, ^{222}Rn concentration in all water resources lower than the reference limit of drinking water as recommended by WHO (100 Bq/l) except water from hot spring that collected at sampling location MC19 in Cherana Putih. However, the concentration for those samples water that below the reference limit of WHO were considered safe for daily consumption.

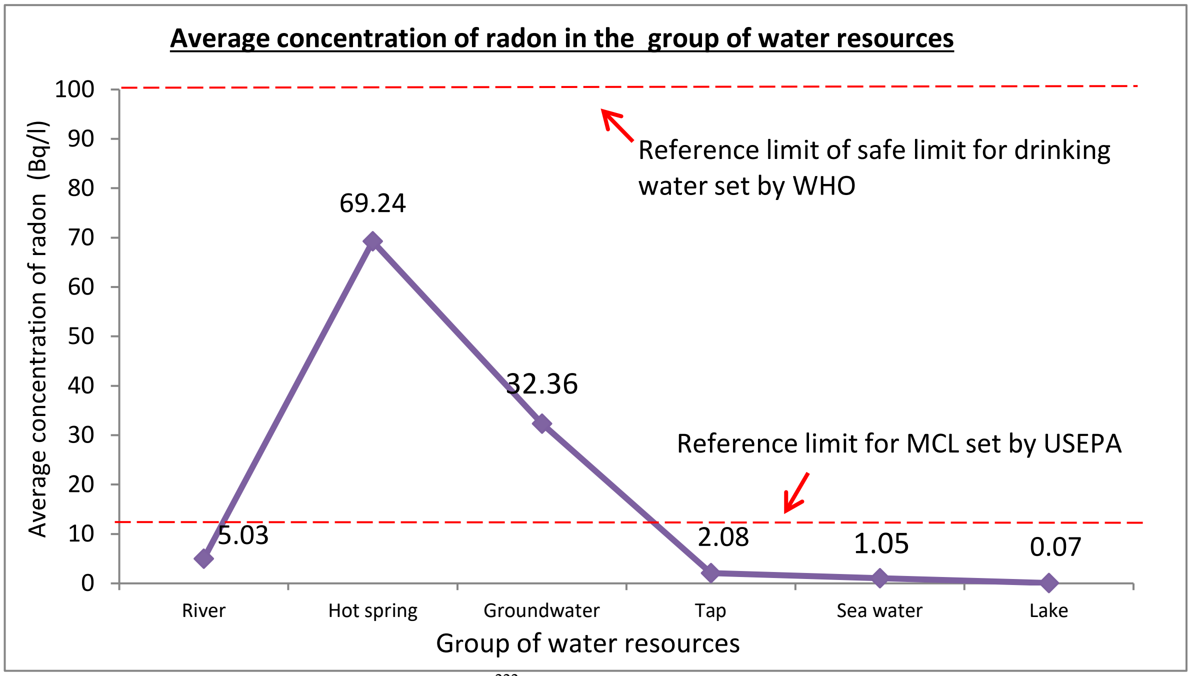

FIGURE 3. The graph of average ^{222}Rn concentration in the group of water resources.

Moreover, the ^{222}Rn concentration for hot spring water and groundwater in this present study were compared to ^{222}Rn activity values from other parts of the world as shows in Table 4 and Table 5, respectively. In Table 3, the average ^{222}Rn concentration in hot spring obtained in the present study is 69.24 ± 3.78 Bq/l, this value revealed that a Malacca level is lower than the measured result in Thailand. Elsewhere, the studies in Indonesia (E.D. Nugraha *et al*. 2021), Jordan (A.T. Al-Kazwini and M.A. Hasan, 2003), Iran (S.M. Mirhosseini, 2016), Perak (H. Nuhu *et al.* 2020), Venezuela (A. Horvarth, 2000) and China (G. Song *et al*. 2002) report ^{222}Rn concentration in hot spring water ranging from 0.04 to 560 Bq/l.

TABLE 4. ^{222}Rn concentrations in hot spring water from other countries as compared to present study

Study area	**^{222}Rn concentration in water (Bq/l)**		**References**
	Range	**Average**	
Suan Phueng district, Ratchaburi province, Thailand.	2 to 154	83.67 ± 1.17	P.Sola *et al.* 2013
West Java region, Indonesia	0.26 to 31	9 ± 1	E.D. Nugraha *et al.* 2021
Jordan	63 to 81	-	A.T. Al-Kazwini and M.A. Hasan, 2003

Iran	9.87 to 46.3	-	S.M. Mirhosseini, 2016
Perak, Malaysia	0.04 ± 0.08 to 0.62 ± 0.32	0.30 ± 0.21	H. Nuhu *et al.* 2020
Northern region of Venezuela	1 to 560	-	A. Horvarth *et al.* 2000
Guangdong, China	53.4 to 292.5	53.85 ± 15.38	G. Song *et al*. 2002
Malacca, Malaysia	8.05 to 111.35	69.24 ± 3.78	Present study

In Table 5, the average ^{222}Rn concentration in groundwater present in this study is lower than the value measured in Gadau Bauchi State, Nigeria. Moreover, the comparison was obviously shows that the average ^{222}Rn concentration in Malacca is higher than those recorded in China (W. Tan *et al*. 2019), Ghana (I. Opoku-Ntim *et al.* 2019), Jamaica (L. Smith and M. Voutchkov, 2017), Iran (M. Malakootian and Y.S. Nejhad, 2017), India (Rangaswamy *et al*. 2016), Iraq (A.A. Ibrahim *et al*. 2017) and Saudi Arabia (E.H. El-Araby *et al.* 2019). In groundwater, the presence of ^{222}Rn is predominantly due to the decay of radium found from the formations of rock and soils at their area (I. Opoku-Ntim *et al*. 2019).

TABLE 5. ^{222}Rn concentrations in groundwater from other countries as compared to present study

Study area	**^{222}Rn concentration in water (Bq/l)**		**References**
	Range	**Average**	
Chenzhou, southern Hunan Province, China	1.29 to 31.31	10.47	W. Tan *et al*. 2019
Obuasi Municipality, Ghana	-	0.09 ± 0.01	I. Opoku-Ntim *et al.* 2019
Offinso Municipality, Ghana	-	0.13 ± 0.04	I. Opoku-Ntim *et al.* 2019
St. Catherine, Jamaica	11 to 41	18 ± 2	L. Smith and M. Voutchkov, 2017
Bam County, Kerman Province, Iran.	1.2 to 9.88	-	M. Malakootian and Y.S. Nejhad, 2017
Gadau Bauchi State, Nigeria	4.92 to 82.89	38.3	H.K. Shu'aibu *et al.* 2020
Shimoga District, India	3.10 ± 0.25	13.60 ± 1.12	Rangaswamy *et al.* 2016

Kirkuk governorate, Iraq	0.108 to 5.630	2.316	A.A. Ibrahim *et al.* 2017
Jazan, Saudi Arabia	2.18 to 5.40	2.95 ± 0.22	E.H. El-Araby *et al.* 2019
Malacca, Malaysia	7.42 to 89.12	32.36 ± 1.84	Present study

Therefore, the various ^{222}Rn concentrations in water resources were closely related to geological rock types, which the geological formation in Malacca is Ordovician Silurian and acid intrusive rock (H.T. Gabdo *et al*. 2015). The ^{222}Rn concentration in water is might be affected by uranium and radium contain in the rocks. Also, the geological structure, porosity of the soil, meteorological parameters can also be a contributory factor for the levels of ^{222}Rn in water resources (I. Opoku-Ntim *et al*. 2019). The result obtained for ^{222}Rn concentration in water in this present study can be used to calculate the AED by using the equation by UNSCEAR. The AED is the activity that enters the respiratory or gastrointestinal tract from the environment that specifically from the intakes of ^{222}Rn in drinking water (P. Ravikumar and R.K. Somashekar, 2016) In the present study, the AED received from ingestion and inhalation of water that containing ^{222}Rn for age groups of infant, children and adult is shows in Table 6**.**

TABLE 6. The total AED for groups of infant, children and adult.

Sampling code	**AED_{ing} ($mSv\ y^{-1}$)**			**AED_{inh} ($mSv\ y^{-1}$)**	**Total AED ($mSv\ y^{-1}$)**		
	Infant	**Children**	**Adult**		**Infant**	**Children**	**Adult**
MC3	0.006	0.002	0.003	0.003	0.009	0.005	0.006
MC5	0.467	0.172	0.226	0.223	0.690	0.395	0.448
MC 7	0.076	0.028	0.037	0.036	0.112	0.064	0.073
MC8	0.020	0.007	0.010	0.010	0.030	0.017	0.019
MC10	0.039	0.014	0.019	0.019	0.058	0.033	0.038
MC12	0.004	0.001	0.002	0.002	0.005	0.003	0.003
MC14	0.007	0.003	0.004	0.003	0.011	0.006	0.007
MC15	0.008	0.003	0.004	0.004	0.012	0.007	0.008
MC16	0.023	0.008	0.011	0.011	0.034	0.019	0.022
MC17	0.043	0.016	0.021	0.020	0.063	0.036	0.041
MC18	0.051	0.019	0.025	0.024	0.075	0.043	0.049

MC19	0.589	0.217	0.284	0.281	0.870	0.497	0.565
MC20	0.098	0.036	0.047	0.047	0.145	0.083	0.094
MC21	0.004	0.001	0.002	0.002	0.006	0.003	0.004
MC22	0.0004	0.0001	0.0002	0.0002	0.001	0.0003	0.0004
MC23	0.012	0.004	0.006	0.006	0.018	0.010	0.012
MC28	0.471	0.174	0.228	0.225	0.696	0.398	0.452
Average	0.113	0.042	0.055	0.054	0.167	0.095	0.108

The AED_{ing} varied from (0.0004 to 0.589) mSv y^{-1}, (0.0001 to 0.217) mSv y^{-1} and (0.0002 to 0.284) mSv y^{-1} with an average value of 0.113 mSv y^{-1}, 0.042 mSv y^{-1} and 0.055 mSv y^{-1} for group of infant, children and adult, respectively. While the AED_{inh} were varied from (0.0002 to 0.281) mSv y^{-1} with an average value of 0.054 mSv y^{-1}.

Furthermore, the total AED through the ingestion and inhalation for age group of infant, children and adult were varied from (0.001 to 0.87) mSv y^{-1} with an average value of 0.167 mSv y^{-1}, (0.0003 to 0.497) mSv y^{-1} with an average value of 0.095 mSv y^{-1}, (0.0004 to 0.565) mSv y^{-1} with an average value of 0.108 mSv y^{-1}, respectively. Based on the calculation, the total AED for infant is significantly higher compared to group of children and adult. The higher AED in infant group is because of the factor difference in metabolism rates and their smaller organ weights that resulting in higher doses for many radionuclides (V. Duggal *et al.* 2020).

Furthermore, the average total AED obtained in this study also was compared to the values recommended by WHO, IAEA and ICRP. The results shows that group of infant and adult have been exceed 0.1 mSv y^{-1} as the value recommended by WHO and IAEA except for group of children. The average total AED for all groups of infant, children and adult were still below 1 to 20 mSv y^{-1} and 10 mSv y^{-1} as the bench mark for ^{222}Rn exposure as recommended by ICRP. In this study, the result obtained was not indicate to any serious hazardous that need an immediate action taken. This is because, there is still having a necessary treatment to carry out to reduce the intake of ^{222}Rn in water sources at the sampling location. In example, it is recommended to treat the water sources by aeration process before supply to domestic use (W.A. Abdurabu *et al.* 2016). Besides that, the sources of water in the study area especially in sampling location MC19 should be boiled in order to release the dissolved ^{222}Rn before drinking (W. Tan *et al*. 2019). For health effect, that recommendation is very helpful as effort to reduce the risk of contracting lung or stomach cancer if in the situation that ^{222}Rn content in drinking water is too high

CONCLUSION

The concentration of ^{222}Rn in 17 sample of water collected in Malacca, Malaysia has a range of (0.07 ± 0.02 to 111.35 ± 2.76) Bq/l, with an average is 21.34 ± 1.51 Bq/l. The estimation of total AED from the intakes of ^{222}Rn in water for group of infant, children and adult were in ranges from (0.001 to 0.87) mSv y^{-1} with an average value of 0.167 mSv y^{-1}, (0.0003 to 0.497) mSv y^{-1} with an average value of 0.095 mSv y^{-1} and (0.0004 to 0.565) mSv y^{-1} with an average value of 0.108 mSv y^{-1}, respectively. The AED for group of infant and adult were exceed the limit recommended by WHO and IAEA. While compared to the ICRP, the AED for all group of ages in this study were recorded below their recommendation limit. However, the data in this study could serve as a reference for any future radiological study especially for ^{222}Rn concentration in water resources in Malacca, Malaysia.

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