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Determination of radon concentration and heavy metals (Ni, Pb, Cd, As, Cr) in drinking and irrigated water sampled from Kulim, Malaysia

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ABSTRACT

Background*:** The presence of heavy metals and radon in water are considered as a potential threat to public health. Modern day industrialization has led to increase pollutants in ecosystem whilst the main source of public radiation exposure, especially exposure from radon, is caused by portable water. This study aimed to estimate the health risk related to radon concentration and heavy metals in drinking and irrigated water. ***Materials and Methods*:** Water samples, from different sources, were collected from Kulim, Malaysia and analyzed for radon and heavy metals using RAD7 and Atomic Absorption Spectrometry. ***Results: Results showed that the maximum average value of radon concentration (16.06±1.7) Bq/l was found in well water used for drinking and irrigation. Minimum average value of radon concentration (2.65±0.33) Bq/l was found in tap water used for drinking purpose. From the measured radon concentrations, age dependent associated annual effective doses and contribution of radon in drinking water to indoor air was calculated which were found below the lower limit of recommended action level. Metal concentrations were found higher in stream and lake waters used for irrigation, compared to well and tap waters. The average daily intake of drinking water and health risk indices were found higher for infants and lower for children and adults. ***Conclusion:*** Radon concentrations in well, stream and lake waters were found higher than EPA recommended level and lower than WHO action level while the annual effective doses and level of heavy metals in water reported in this study were found lower than recommended levels.

Keywords: *Water, radon, heavy metals, annual effective dose, RAD7.*

INTRODUCTION

The main sources of radiation exposure are primordial, cosmic and artificial radiations. About radiation hygiene, radon and its progeny are the most important factors of the public exposure, which occupy from the sources of natural radiation more than half of the dose exposures accessing the general public [(1)]. ^{222}Rn is the most important isotope of concern to public. It has a half-life of 3.82 days, emits 5.5 MeV alpha particles, and generate radioactive progenies. Radon and its short-lived decay products constitute about 53% of the total dose originating from natural sources [(2)]. Ground water can bring additional radon into dwelling and workplaces, increasing health risk. Radon can easily escape from water into air, when the water is utilized for cleaning, showering and other daily life routines. Radon in drinking water can increase the indoor radon concentration level in homes by escaping from water supply [(3)].

Radon in drinking water contributes inhalation and ingestion doses. Alphas from ^{222}Rn can directly interact with tissues in the body, and could damage DNA and RNA structures. In general, the levels of radon concentration in surface water are very low [4], while the highest levels of radon concentration are reported in ground water flowing through granitic sand, granite and gravel formations which ranges from 1 to 50, 100 to 300 and 100 to 50,000 kBq/m^3 in aquifer and sedimentary rocks, deep wells and crystalline rocks, respectively [2-6].

Materials such as gravel, sand and bedrocks, inside the earth, may contains heavy metals, which are absorbed and dissolved by water that we take from different sources for irrigation and domestic purposes [7-9]. Rapid urbanization, and discordant industrial development, in different parts of world, has affected the ecosystem, food safety, and present potential threats to human health. Irrigation of agriculture soil through waste water results in soil contamination and ultimately affects food quality. On the other hand industrial wastes are sources of trace amount of heavy metals that are being discharged into the aquatic environment and pose threat to human health through drinking water and food. One of the important issues is heavy metals contamination, such as Cd, Cr, Ni, Pb and As, in ground water due to their toxicity even at low concentration [10]. Cadmium is known as carcinogenic and considered a cause of different types of cancers. Most physiological effects such as anemia, hypertension, poor mineralization and damage to renal tubules are encountered due to exposure of cadmium [11]. High quantity of chromium affects the lipid and sugar metabolism in humans and its deficiency is considered as leading cause of disease called chromium deficiency [12]. Nickel is an essential trace element in animals and humans bodies. It is associated with the molecules of RNA and DNA and is considered as a regulatory element for different enzyme system. Lead is known as a cause of several damages in human kidneys, reproductive system, brain and nervous system [13]. Monitoring of quality of drinking and irrigated water is one of the most important part of public health studies.

The present study aimed to estimate the health risk related to radon concentration and heavy metals in drinking and irrigated water of Kulim, Malaysia by estimating considerable activities of humans like agriculture and industrial activities. The results obtained from this study will contribute to a database of environmental radioactivity measurements and will be useful for assessing the safety of the drinking and irrigated water.

MATERIALS AND METHODS

Samples collection

The drinking water sources in the study area are tap and well water. Irrigation of agriculture soil is carried by well, stream and lake water. Water samples were collected from streams, lakes, wells and tap in a 1.5 liter clean polyethylene bottles from 18 locations of Kulim, Kedah, Malaysia. Bottles were pre-washed with 15% nitric acid and three times with double de-ionized water. Water samples from wells were collected directly from their locations after purging for 10 minutes to ensure sample quality. Tap water was collected using plastic funnel and water samples from streams were taken within 5 cm of the water surface and capped right inside the water body. All samples were labeled by sample code, date and time of collection. Samples were stored in Medical Physics Lab, School of Physics, Universiti Sains Malaysia. The sampling sits are listed in table 1. Figure 1 shows the map of study area and sites locations.

Measurement of radon concentration and heavy metals

In the current study, for the measurement of radon in drinking water, we have used a multipurpose device called RAD7 [14]. It is considered a comprehensive system for measurement of radon in water, soil, and air [15].

RAD7 is a solid state, ion implanted, planer, silicon alpha detector which convert alpha radiation to an electrical signal. It has internal sample cell of 0.7 dm^3 (0.7l) hemisphere, coated internally with electrical conductor. Radon decays within the cell by leaving it's positively charge ion ^{218}Po which is propagated by electric field towards detector. When the short-lived polonium-218 nucleus decays upon the detector's active surface, its alpha particle has a 50% probability of entering the detector and producing an electrical signal proportional in strength to the energy of the alpha particle. Subsequent decays of the same nucleus produce beta particles, which are not detected, or alpha particles of different energy. Different isotopes have different alpha energies, and produce different strength signals in the detector.

Radon concentration in water was measured by calibrated alpha spectrometer RAD7 Model 2890 according to the EPA Protocol test [16]. In calibration, the RAD7 is exposed to a known concentration of radon (or thoron) and the count rates are measured. Thirty minutes measuring time at Wat-250 protocol and Grab mode was taken for all samples. Before analysis, sample of 250 ml each was sealed off for 3-4 hours. The concentration of radon measured by RAD7 in pCi/L was converted in to Bq/l using relation, 1pCi = 0.037 Bq.

Contribution of radon concentration in water to indoor was calculated using equation (1) [17].

$$C_{aRn}{}^{222} = C_{wRn}{}^{222} \times W \times \frac{e}{(V \times \lambda_c)} \quad (1)$$

Where $C_{aRn}{}^{222}$ is the contribution of radon in water to indoor (Bq/l), $C_{wRn}{}^{222}$ is the radon concentration of water (Bq/l), W is the water consumption (0.01 m^3/h per person), V is the bulk of indoor room (20 m^3 per person), e is the coefficient of radon transfers from dosmistic water to indoor air (0.5) and λ_c is air exchange rate (0.7 h^{-1}) [18,19]. Annual effective dose for ingestion was calculated using equation (2) [20,21].

$$E_d = A_c \times A_i \times C_f \quad (2)$$

Where E_d is annual effective dose for ingestion, A_c is radon concentration (Bq/l), A_i is the intake of water (230, 330 and 730 l/y for babies, childern and adult, respectively) [22] and C_f is dose conversion factors for radon (3.5, 5.9 and 23 nSv Bq^{-1} for audlts, childerns and infants, respectively) [23].

For assessment of heavy metals, all samples of water were filtered through double rings filter paper and concentrated from 1000 ml to 50 ml with 5 ml nitric acid to dissolve the metals. Atomic Absorption Spectrometry (Perkin-Elmer, Model Analyst 200, Shimadzu, Model AA-700) was used for analysis of Cd, Ni, Cr, Pb and As. For accuracy assurance, each sample was analyzed in triplicate and instrument was calibrated continuously with the certified standard solution 0f 1000 ppm (Merck Darmstadt, Germany and Fisher Scientific, UK limited) [24].

The average metal daily intake of drinking water for infants, children and adults were estimated by using equation (3) [25].

$$MDI = \frac{C_m \times D_{wi}}{B_{aw}} \quad (3)$$

Where MDI is metal daily intake (mg/kg/day) C_m, represents concentration of heavy metals in drinking water, D_{wi} shows average daily intake and B_{aw} average body weight (6.8 Kg) [26], (32Kg) [27] and (62.65kg)[28] for infants, children and adults, respectively.

To estimate non-carcinogenic risk, Health risk indices (HRI) were evaluated from the intake of water for Cr, Ni, Cd, As and Pb using equation (4) [29].

$$HRI = \frac{MDI}{RfD} \quad (4)$$

Where HRI is health risk indices, MDI is metal daily intake and RfD is the reference dose. The values of RfD for Ar, Cr, Ni, Cd and Pb were 0.0003, 1.5, 0.02, 0.001 and 0.004 mg/kg bw/ day, respectively [30, 31].

RESULTS

The level of radon concentration in water and its contribution to indoor air are presented in table 2. The values of radon concentration in well water varied from 10 to 20 Bq/l with the average value of 16.06 Bq/l, while in case of lake, stream and tap water its values ranged from 10.9 to 11.6 Bq/l with the average value of 11.25 Bq/l, 7.2 to 11.9 Bq/l with the average value of 9.74 Bq/l and 1.4 to 4.7 Bq/l with the average value of 2.65 Bq/l, respectively. The contribution of radon in water to indoor air was ranged from 0.5 to 7.14 Bq/m^3. Age dependent annual effective dose due to ingestion of radon in drinking water is reported in table 3. The annual effective doses in drinking water varied from 0.007 to 0.106 mSv/y, 0.003 to 0.039 mSv/y and 0.003 to 0.045 mSv/y for infants, children and adults, respectively.

Table 1. Locations of sampling sites.

S. No.	Site Name	Site Code	Source of water	Coordinates
1	Kampung Sg Selaung Atas	K	Well	N 05° 26' 591″ E 100° 32' 611″
2	Kampung Guar Lobak	K6	Well	N 05° 31' 514″ E 100° 32' 326″
3	Kampung Ladong Bukit Padang Sarai	K10-2	Well	N 05° 28' 947″ E 100° 35' 884″
4	Kampung Sentosa	K11	Well	N 05° 31' 408″ E 100° 32' 811″
5	Kampung Sentosa	K11-2	Well	N 05° 31' 275″ E 100° 35' 494″
6	Kampung Batu 4 Sg Ular	K4	Stream	N 05° 21' 052″ E 100° 34' 954″
7	Kampung Bagar Musah	K5	Stream	N 05° 28' 308″ E 100° 37' 783″
8	River sungai Jarak Padong Saria	K12	Stream	N 05° 28' 489″ E 100° 32' 604″
9	River Sungai Binjanl	K16	Stream	N 05° 17' 968″ E100° 32' 025″
10	River Sungai Kulim	K17	Stream	N 05° 23' 288″ E100° 33' 613″
11	Kulim Perdana	K3	Lake	N 05° 24' 052″ E 100° 34' 546″
12	Kg Tasek Bandran	K14	Lake	N 05° 39' 6.74″ E 100° 28' 59.5″
13	Kampung Pondok Labu	K7	Tap	N 05° 23' 066″ E 100° 31' 487″
14	Kampung Sungai Seluang	K8	Tap	N 05° 27' 441″ E 100° 32' 806″
15	Kampung Permatang Durian	K9	Tap	N 05° 29' 117″ E 100° 32' 635″
16	Kampung Guar Lobak	K10	Tap	N 05° 32' 097″ E 100° 32' 589″
17	Kampung Simpung Tiga Keladi	K13	Tap	N 05° 23' 013″ E 100° 33' 102″
18	Kampung Permatang Tiong	K15	Tap	N 05° 17' 082″ E 100° 32' 169″

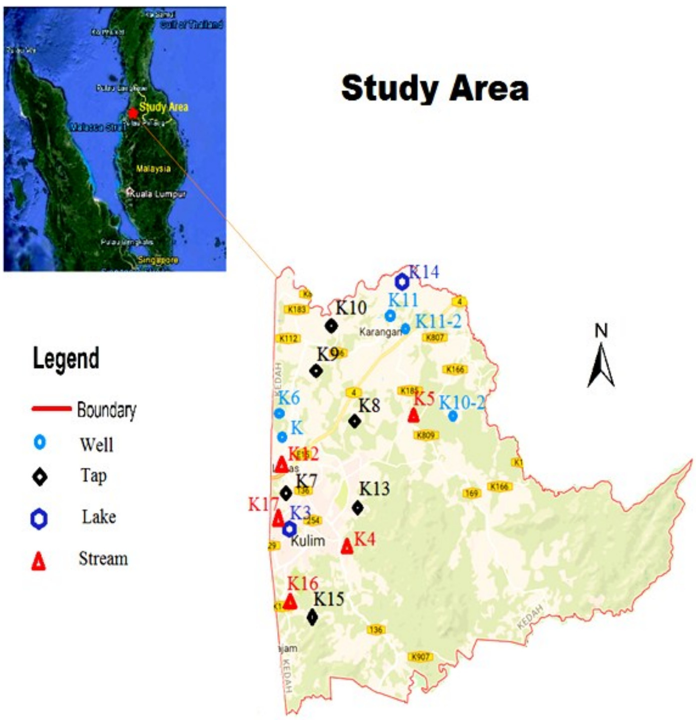

Figure 1. Map of study area and location of sampling sites (source Google Earth).

Table 2. Radon concentration and its contribution to indoor from drinking and irrigated water.

S No	Sample Code	Source of water	Radon Concentration in water (Bq/l)	Contribution of radon to indoor C_A ^{222}Rn (Bq/m^3)
1	K	Well	1.5 ± 16.1	0.35 ± 5.75
2	K6	Well	1.9 ± 19.6	0.67 ± 7.00
3	K10-2	Well	2.2 ± 20.0	0.78 ± 7.14
4	K11	Well	1.7 ± 14.4	0.60 ± 5.14
5	K11-2	Well	1.2 ± 10.2	0.42 ± 3.64
6	K4	Stream	0.90 ± 7.2	0.32 ± 2.57
7	K5	Stream	0.87 ± 8.2	0.31 ± 2.93
8	K12	Stream	0.93 ± 10.2	0.33 ± 3.64
9	K16	Stream	1.09 ± 11.9	0.38 ± 4.25
10	K17	Stream	1.16 ± 11.2	0.41 ± 4.00
11	K3	Lake	1.14 ± 11.6	0.4 ± 4.14
12	K14	Lake	1.11 ± 10.9	0.39 ± 3.89
13	K7	Tap	0.27 ± 1.4	0.096 ± 0.5
14	K8	Tap	0.34 ± 3.0	0.12 ± 1.07
15	K9	Tap	0.34 ± 4.7	0.12 ± 1.68
16	K10	Tap	0.49 ± 2.4	0.17 ± 0.85
17	K13	Tap	0.35 ± 1.8	0.12 ± 0.64
18	K15	Tap	0.21 ± 2.6	0.75 ± 0.92

Table 2. Radon concentration and its contribution to indoor from drinking and irrigated water.

S No	Sample Code	Source of water	1-0years	16-2years	17years
1	K	Well	0.007 ± 0.085	0.003 ± 0.031	0.003 ± 0.036
2	K6	Well	0.01 ± 0.104	0.004 ± 0.038	0.004 ± 0.044
3	K10-2	Well	0.011 ± 0.106	0.004 ± 0.039	0.005 ± 0.045
4	K11	Well	0.008 ± 0.076	0.003 ± 0.028	0.004 ± 0.032
5	K11-2	Well	0.006 ± 0.054	0.002 ± 0.02	0.003 ±0.023
6	K7	Tap	0.001 ± 0.007	0.0005 ± 0.003	0.0006 ± 0.003
7	K8	Tap	0.001 ± 0.016	0.0007 ± 0.006	0.0008 ± 0.007
8	K9	Tap	0.001 ± 0.024	0.0007 ± 0.009	0.0008 ± 0.001
9	K10	Tap	0.002 ± 0.012	0.001 ± 0.005	0.001 ± 0.005
10	K13	Tap	0.001 ± 0.009	0.0007 ± 0.003	0.0008 ± 0.004
11	K15	Tap	0.001 ± 0.013	0.0004 ± 0.005	0.0005 ± 0.006

Table 4 presents the concentration of heavy metals in drinking and irrigated water (32-34). The maximum concentration of heavy metals were found (11.8 ± 1.16) for Ni, (8.72 ± 1.08) for Pb, (4.66 ± 0.66) for Cd, (5.4 ± 0.68) for As and (2.52 ± 0.24) μg/l for Cr in stream water used for irrigation and minimum were found below detection limit for Cr, (0.24 ± 0.14) for As, (0.28 ± 0.08) for Ni, (0.52 ± 0.08) for Cd and (1.14 ± 0.12) μg/l for Pb in tap water used for drinking in the study area. The order of distribution of heavy metals were found at the pattern Ni > Pb > Cd > As > Cr. Two samples (K8, K9) have Cr concentration below the detection limit. Daily average intake and health risk indices are summarized in Table 5. The values of daily average intake (MDI) and health risk indices (HRI) due to As, Cr, Ni, Cd and Pb in drinking water followed the pattern infants > children > adults.

Table 4. Heavy metals in drinking and irrigated water (μg/l) and international standard limits.

S No	Site Code	Source of water	As	Cr	Ni	Cd	Pb
1	K	Well	0.1 ± 1.5	0.16 ± 1.42	0.14 ± 1.64	0.12 ± 1.12	0.24 ± 2.36
2	K6	Well	0.08 ± 0.9	0.08 ± 0.46	0.78 ± 6.02	0.14 ± 1.62	0.9 ± 7.8
3	K10-2	Well	0.12 ± 0.84	0.12 ± 0.62	0.58 ± 4.0	0.56 ± 3.78	0.7 ± 4.8
4	K11	Well	0.14 ± 1.92	0.08 ± 0.3	0.48 ± 3.6	0.28 ± 2.64	0.28 ± 2.14
5	K11-2	Well	0.22 ± 1.7	0.14 ± 0.74	0.66 ± 4.6	0.14 ± 1.38	0.52 ± 3.78
6	K4	Stream	0.3 ± 2.18	0.24 ± 2.52	0.92 ± 6.58	0.26 ± 2.56	1.08 ± 8.72
7	K5	Stream	0.68 ± 5.4	0.18 ± 1.18	0.46 ± 3.42	0.3 ± 2.04	0.76 ± 4.72
8	K12	Stream	0.5 ± 3.24	0.14 ± 0.74	1.08 ± 10.0	0.4 ± 2.78	1.0 ± 7.38
9	K16	Stream	0.3 ± 2.28	0.14 ± 1.34	0.68 ± 4.38	0.54 ± 3.86	0.32 ± 2.28
10	K17	Stream	0.32 ± 2.46	0.12 ± 1.1	1.16 ± 11.8	0.66 ± 4.66	0.84 ± 6.42
11	K3	Lake	0.24 ± 2.04	0.02 ± 0.18	0.9 ± 9.14	0.62 ± 3.68	0.58 ± 3.64
12	K14	Lake	0.48 ± 3.62	0.16 ± 1.68	0.84 ± 5.7	0.2 ± 0.88	0.8 ± 5.5
13	K7	Tap	0.2 ± 1.6	0.02 ± 0.12	0.44 ± 3.06	0.18 ± 1.52	0.48 ± 3.52
14	K8	Tap	0.06 ± 0.82	BDL[d]	0.12 ± 1.06	0.01 ± 0.6	0.52 ± 4.6
15	K9	Tap	0.14 ± 0.24	BDL[d]	0.32 ± 2.76	0.4 ± 2.82	0.12 ± 1.14
16	K10	Tap	0.12 ± 0.58	0.08 ± 0.14	0.8 ± 5.94	0.08 ± 0.52	0.52 ± 3.08
17	K13	Tap	0.16 ± 1.02	0.14 ± 0.24	0.16 ± 1.96	0.36 ± 2.56	0.3 ± 2.1
18	K15	Tap	0.16 ± 0.96	0.06 ± 1.2	0.08 ± 0.28	0.16 ± 1.34	0.8 ± 5.92
Mean			0.24 ± 1.85	0.1 ± 0.77	0.59 ± 4.77	0.31 ± 2.35	0.56 ± 4.43
WHO[a]			10	50	70	3	10
USEPA[b]PCD[c]			10[b]	100[b]	20[c]	5[b]	15[b]

dBelow detection limit, a(WHO 2011), b(EPA 2012), c(PCD 2004) (32-34)

Table 5. Daily average intake (mg/kg/day) and health risk indices from drinking water.

S No	Individual		As	Cr	Ni	Cd	Pb
1	Infants	MDI	10^{-4}×(0.12 ± 1.0)	10^{-5}×(0.73 ± 4.3)	10^{-4}×(0.38 ± 2.9)	10^{-4}×(0.17 ± 1.3)	10^{-4}×(0.44 ± 3.4)
		HRI	(0.04 ± 0.33)	10^{-5}×(0.48 ± 2.8)	(0.002 ± 0.014)	(0.017 ± 0.13)	(0.011 ± 0.084)
2	Children	MDI	10^{-5}×(0.38 ± 3.0)	10^{-5}×(0.22 ± 1.3)	10^{-5}×(0.11 ± 9.0)	10^{-5}×(0.54 ± 4.0)	10^{-5}×(0.13 ± 9.3)
		HRI	(0.012 ± 0.1)	10^{-6}×(1.4 ± 8.6)	10^{-3}× (0.05 ± 4.5)	(0.005 ± 0.04)	(0.0003 ± 0.023)
3	Adults	MDI	10^{-5}×(0.42 ± 3.4)	10^{-5}×(0.24 ± 1.4)	10^{-4}×(0.12 ± 1.0)	10^{-5}×(0.6 ± 4.4)	10^{-4}×(0.15 ± 1.1)
		HRI	(0.014 ± 0.11)	10^{-6}×(1.6 ± 9.3)	10^{-3}× (0.6 ± 5.0)	(0.006 ± 0.044)	(0.004 ± 0.027)

DISCUSSION

The maximum value of radon concentration found in well water collected from Kumpung Ladong Bukit Padang Sarai is attributed to 150 feet depth of well, as the concentration of radon is usually high in ground water due to high concentration of uranium and radium contents [35] and lower was found in tap water which may be due to filtration treatment of water in the study area. The radon concentration in streams and lakes were found comparatively higher than the tap water due to dissolved radioactive materials. The risk associated with radon concentration is typically low compared with total inhaled radon but cannot be neglected because exposure occurs through both inhalation of released radon and consumption of dissolved gas and its radioactive progenies from the water. The concentration levels of radon in seven samples collected from wells (sample no. K, K6, and K10-2 K11), streams (sample no. K16, K17) and lakes (sample no. K3) were found higher than the recommended level of United States Environmental Protection Agency but found lower than the recommended level of WHO [36-38]. Table 6 [39-44] shows the comparison of radon concentration in the well and tap waters with the results of different studies conducted around the world. It can be seen that for the well and tap waters the radon concentration obtained for current study was higher than reported values of all the countries except Iran. The higher concentration of radon in water may be due to number of factors, e.g. the geological structure, meteorological parameters, providing surface reservoirs, etc.

The results tabulated in Table 2 shows that 1 Bq/l (1 kBq/m^3) of radon in drinking water can produce an accessorial indoor concentration of radon of 3.57×10^{-4} (0.357 Bq/m^3). The average annual effective doses were found below the permissible limit of 1 mSv/y [36, 23].

Concentrations of heavy metals were found higher in rivers and lakes water as compared to tap and well water. This variation is attributed to human activities which includes the rapid urbanization especially construction for housing estate, the ongoing 145 km^2 Kulim Hi-Tech Industrial park, sand mining activity [43]. But all values of heavy metals in Kulim area were found lower than the recommended limits as shown in.

The results of HRI assessment indicate that water of the study area is safe for drinking as the contamination of heavy metals are low. HRI were found less than 1, therefore, no health risk via consumption of water is expected.

Table 6. Comparison of radon concentration in water with previous work.

S.No.	Radon Concentration (Bq/l)		Methodology	Country	Reference
	Well	Tap			
1	1.21 ± 0.005	0.602 ± 0.003	CR-39	Pakistan	(39)
2	2.050	0.174	RAD7	Iraq	(40)
3	21.77	11.44	PRASSI	Iran	(41)
4	13.76	n.a	RAD7	Turkey	(42)
5	n.a	0.025-0.128	AlphaGUARD PQ2000 PRO	Turkey	(43)
6	1.88-43.01	1.23-11.94	RAD7	Iran	(44)
7	16.02 ± 1.7	2.64 ± 0.33	RAD7	Malaysia	Present Study

n.a., not available

CONCLUSION

To conclude, the present study invisteged the public health risk, of Kulim population. Malaysia, from exposure of radon and heavy matels presence in drinking and irrigated water. High radon concentrations were observed in well water sources. Average radon concentration in water collected from rivers and Lakes, used for irrigation, were found higher then tap water. It is recommended to boil the well water before use for drinking in oder to decrease the level of radon concentration. The results reveal the fact that values of annual effective doses due to radon exposure are below the permissable limit proposed by UNSCEAR and WHO. Presence of metals with different sources of waters were found below the recommended levels.

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