Radon-222 from different sources of water and the assessment of health hazard

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ABSTRACT

Water samples collected from different sources were analysed for radon concentrations in order to evaluate the health effect associated with radon in water. The radon concentrations were in the range of 3.56–98.57, 0.88–25.49, 0.73–1.35 and 0.24–1.03 $Bq.L^{-1}$ for borehole, well, packaged and utility water, respectively. Samples from boreholes had the highest radon concentrations with about 67% being higher than the threshold value of 11.1 $Bq.L^{-1}$ recommended by the USEPA. The mean annual effective dose (AED) due to ingestion for adult, child and infant ranged from 8.71×10^{-3} to 0.831 $mSv.y^{-1}$ for the different sources. The mean AED calculated for consuming water from boreholes and wells for the three age groups were higher than the recommended reference dose level of 0.1 $mSv.y^{-1}$. The mean AED due to inhalation of radon in drinking water was negligible, ranging from 0.13 to 6.20 $\mu Sv.y^{-1}$. The health burden associated with radon in water in the study is through ingestion of water directly from boreholes.

Key words | activity concentration, annual effective dose, ingestion, inhalation, radon, water

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INTRODUCTION

There are three naturally occurring radioactive decay series in the Earth crust originating from ^{238}U, ^{235}U and ^{232}Th. Each of the series has an isotope of radon (symbol: Rn); ^{222}Rn ($T_{1/2} = 3.8235$ days), ^{219}Rn ($T_{1/2} = 3.96$ seconds) and ^{220}Rn ($T_{1/2} = 0.9267$ minutes) from the ^{238}U-, ^{235}U- and ^{232}Th-decay series, respectively. ^{219}Rn and ^{220}Rn are short-lived nuclides and as such they are not abundant in nature (Dewayne & Gesell 1992). ^{222}Rn can reach secular equilibrium in ca. 25 days with its parent element ^{226}Ra contained in rocks (Andrews & Lee 1979) and thus it can accumulate in appreciable amounts in groundwater (Kendall & McDonnell 1998; Wu *et al.* 2003). Radon is odourless, colorless and tasteless, thereby making it difficult to detect its presence in water with the human senses. ^{222}Rn decays to ^{218}Po by emitting alpha particles which are a potential health hazard if radon is inhaled or ingested (National Research Council 1999).

The inhalation of radon progeny is the largest single source of radiation exposure to the population, contributing to about 52% of the total dose due to natural radiation (UNSCEAR 2000). A relationship between lung cancer and inhalation of radon decay products has been demonstrated for underground miners (Lubin *et al.* 1995) and in domestic environments (Lubin & Boice 1997). Radon is a factor of stomach radiation burden due to consumption of water (Nikolopoulos *et al.* 2009).

Radon in tap water may lead to exposure from ingestion of drinking water and from the inhalation of radon released to air when water is used for bathing, cooking, washing, etc. (UNSCEAR 2000). Once in the building, water with an elevated level of radon can cause radon to diffuse into the indoor atmosphere and increase the overall radon levels (Appleton 2005). A link between lung cancer and inhalation of radon has been established and ingestion of radon has been weakly linked to stomach cancer (Hopke *et al.* 2000). A high concentration of radon in drinking water causes stomach cancer (Tabassum & Mujtaba 2012; Thabayneh 2015). As such radon has been identified as a public health

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concern when present in all types of drinking water. Due to the health burden of radon, this study is aimed at assessing the radon concentration in different sources of water at the University of Ibadan, Nigeria and evaluating the effective dose due to the ingestion and inhalation of radon in water.

MATERIALS AND METHODS

Sample collection

Forty-six water samples were collected from different sources. The sources include borehole (drilled deep well), well (dung shallow well), treated water from the university utility water supply and packaged water. The concentration of dissolved radon in water is highly dependent on the extent to which the water is aerated. Therefore, in order to measure the actual public exposure levels, water samples were collected at consumer's fetching points.

Water samples were collected using 400 mL glass bottles. For borehole sources, water from the tap was left running for several minutes, a bucket was filled to overflowing and then raised so that the tap is below the water surface. The collection bottle was then immersed and filled from the bottom of the bucket and capped underwater. This was done to prevent air pockets and bubbles. Water from wells was collected with the aid of a bailer and bottles were carefully submerged into the bailer to collect water samples. Each sample was carefully labeled and the time and date of sampling were noted. Treated water (sourced from a surface dam) from the utility water supply of the university was collected from storage tanks in a residential area where the water had been directly supplied from the treatment plant. Packaged water was purchased from an open market in the university. The source of the packaged water is a borehole in the university; however it had been subjected to some processes before packaging. After collection, samples were quickly taken to the laboratory for measurement. Two aliquots were measured for each sample and the mean value was obtained.

Radon measurement

In this study, an AlphaGUARD portable radon monitor manufactured by Saphymo GmbH, Germany, was used for the measurement of radon concentration in water. AlphaGUARD is a potable, battery operated monitor with a high storage capacity. It measures and records radon concentrations simultaneously with the ambient temperature, relative humidity and atmospheric pressure with integrated sensors. AlphaGUARD incorporates a pulse-counting ionization chamber (alpha spectroscopy). It is suitable for the continuous monitoring of radon concentrations between 2 and 2,000,000 Bq/m^3 through the optimal geometry of its chamber and intelligent signal evaluation. AlphaGUARD offers high detection efficiency, a wide measurement range, fast response and permanent, maintenance-free operation with long-term stable calibration. The instrument is insensitive to both high humidity and vibrations. With AlphaPUMP, AquaKIT and the Soil Gas Probe, the equipment is suitable for the measurement of radon in water samples and soil gas. The measurement of radon in water was carried out using the AlphaGUARD via a special unit (AquaKIT). The unit consists of a vessel used for forced degassing of radon diluted in water samples, a security vessel used for water drop deposition. Vessels and AlphaGUARD were connected via plastic radon proof tubes. Forced degassing of radon gas is performed by circulating the air in the set up with the use of a pump (AquaPUMP). A 100 mL water sample was placed in an appropriate system of glass vessels connected to the detector through the air pump, following the recommendations of the manufacturer (Saphymo GmbH 2015). The radon detector, AlphaGUARD, is based on the optimized design of a pulse-ionization chamber. In regular operation this detector measures the radioactivity of the air using the diffusion of gas through the large surface of the glass fiber filter installed inside the ionization chamber. This filter allows only the ^{222}Rn gas to pass through and prevents the products of the radon decay from entering into the ionizing chamber. It also protects the ionizing chamber from contamination by dust particles (Corrêa *et al.* 2011).

RESULTS AND DISCUSSION

Radon concentration

The radon activity concentrations measured in the water samples from the different sources were calculated using the following expression:

$$C_{\text{water}} = \frac{C_{\text{air}}\left(\left(\left(V_{\text{sys}} - V_{\text{samp}}\right)/V_{\text{samp}}\right) + k\right) - C_{bg}}{1000} \quad (1)$$

where C_{water} is the concentration of radon in water in Bq.l^{-1}, C_{air} (Bq.m^{-3}) is the concentration of radon in the air of the system after it has been released from the water sample, C_{bg} is the background radon activity; V_{sys} is the total volume of the measurements circuit (mL) and V_{samp} is the volume (mL) of water sample. The diffusion coefficient k is given as 0.26 in the AlphaGUARD manual (Saphymo GmbH 2015).

Table 1 contains the result of radon concentrations in water samples collected from boreholes. The radon concentration in water ranged from 3.56 ± 0.74 Bq.l^{-1} to 98.57 ± 5.10 Bq.l^{-1} with a median value of 14.63 ± 1.9 Bq.l^{-1}. This is a major source of water in the students' halls of residence. The water is pumped directly from boreholes everyday without any treatment into reserviour tanks where it is taken for drinking, cooking and bathing. It is possible that if the water had been treated, most of the dissolved radon in water would be vented/released during treatment and this might result to low radon concentrations. The radon concentrations in water samples collected from wells are presented in Table 2. The radon activity concentration levels in the well

Table 1 | Radon concentrations and the AED due to ingestion (H_{ing}) and inhalation (H_{inh}) of radon in water samples from boreholes

	Radon concentration		H_{ing} (mSv.y^{-1})			
S/N	C_{air} (Bq.m^{-3})	C_{water} (Bq.L^{-1})	Adult	Child	Infant	H_{inh} (μSv.y^{-1})
1	4,885 ± 311	50.22 ± 3.20	0.825	1.650	0.962	12.13
2	706 ± 113	7.26 ± 1.16	0.119	0.238	0.139	1.78
3	895 ± 106	9.20 ± 1.09	0.151	0.302	0.176	2.26
4	3,045 ± 232	31.30 ± 2.39	0.514	1.028	0.599	7.67
5	7,780 ± 482	79.98 ± 4.96	1.314	2.628	1.533	19.61
6	1,003 ± 101	10.31 ± 1.03	0.169	0.338	0.197	2.53
7	1,520 ± 134	15.63 ± 1.37	0.257	0.514	0.299	3.83
8	4,300 ± 289	44.20 ± 2.95	0.726	1.452	0.847	10.84
9	1,190 ± 113	12.23 ± 1.16	0.201	0.402	0.234	2.99
10	1,325 ± 123	13.62 ± 1.26	0.224	0.448	0.261	3.34
11	845 ± 90	8.69 ± 0.93	0.143	0.286	0.167	2.13
12	1,013 ± 103	10.41 ± 1.06	0.171	0.342	0.199	2.55
13	725 ± 90	7.45 ± 0.93	0.122	0.244	0.143	1.83
14	9,589 ± 496	98.57 ± 5.10	1.619	3.238	1.889	24.16
15	5,155 ± 332	52.99 ± 3.41	0.870	1.740	1.015	12.99
16	2,015 ± 185	20.71 ± 1.90	0.340	0.680	0.397	5.08
17	1,320 ± 116	13.57 ± 1.19	0.223	0.446	0.260	3.33
18	1,230 ± 119	12.64 ± 1.22	0.208	0.416	0.242	3.10
19	2,080 ± 169	21.38 ± 1.74	0.351	0.702	0.410	5.24
20	710 ± 82	7.30 ± 0.84	0.120	0.240	0.140	1.79
21	3,410 ± 228	35.05 ± 2.34	0.576	1.152	0.672	8.59
22	346 ± 72	3.56 ± 0.74	0.058	0.116	0.068	0.87
23	2,280 ± 166	23.44 ± 1.71	0.385	0.770	0.449	5.75
24	1,690 ± 138	17.37 ± 1.42	0.285	0.570	0.332	4.26
Mean	2,461 ± 2,353	25.30 ± 24.19	0.415	0.831	0.485	6.20

Table 2 | Radon concentrations and the AED due to ingestion (H_{ing}) and inhalation (H_{inh}) of radon in water samples from wells

	Radon concentration		H_{ing} (mSv.y^{-1})			
S/N	C_{air} (Bq.m^{-3})	C_{water} (Bq.L^{-1})	Adult	Child	Infant	H_{inh} (µSv.y^{-1})
1	1,285 ± 120	13.21 ± 1.23	0.217	0.434	0.253	3.24
2	1,455 ± 132	14.96 ± 1.35	0.246	0.492	0.287	3.67
3	483 ± 84	4.97 ± 0.86	0.082	0.164	0.095	1.22
4	608 ± 77	6.25 ± 0.79	0.103	0.206	0.119	1.53
5	772 ± 82	7.94 ± 0.84	0.130	0.260	0.152	1.95
6	86 ± 55	0.88 ± 0.57	0.014	0.028	0.017	0.22
7	223 ± 33	2.29 ± 0.34	0.038	0.076	0.044	0.56
8	2,480 ± 187	25.49 ± 1.92	0.419	0.838	0.488	6.25
9	424 ± 64	4.36 ± 0.66	0.072	0.144	0.084	1.07
10	604 ± 66	6.21 ± 0.68	0.102	0.204	0.119	1.52
11	476 ± 49	4.89 ± 0.50	0.080	0.160	0.094	1.19
Mean	809 ± 689	8.31 ± 7.09	0.137	0.273	0.159	2.04

water ranged from 0.88 ± 0.57 Bq.l^{-1} to 25.49 ± 1.92 Bq.l^{-1} with a median value of 6.21 ± 0.68 Bq.l^{-1}. Table 3 presents the results for the packaged water which ranged from 0.73 ± 0.13 Bq.l^{-1} to 1.35 ± 0.30 Bq.l^{-1} with a median value of 0.91 ± 0.19 Bq.l^{-1}. For the utility water supply shown in Table 4, the radon concentration ranged from 0.24 ± 0.11 to 1.03 ± 0.20 Bq.l^{-1} with a median value of 0.46 ± 0.12 Bq.l^{-1}. It is observed that the radon concentrations of the packaged and utility water are much lower than those of the borehole and well even though the packaged water is sourced from a borehole. This could be attributed to the treatment process which could lead to the radon degassing. This study showed a high level of radon in borehole water followed by well water. The utility water had the least amount of radon concentration, which is due to the fact that radon is readily released from surface water. About 67% (borehole) and 27% (well) of the investigated water samples had radon concentrations higher than 11.1 Bq.l^{-1}, recommended by the United States Environmental Protection Agency (EPA 1991; Paschuk *et al.* 2013).

Table 3 | Radon concentrations and the AED due to ingestion (H_{ing}) and inhalation (H_{inh}) of radon in water samples from packaged water

	Radon concentration		H_{ing} (mSv.y^{-1})			
S/N	C_{air} (Bq.m^{-3})	C_{water} (Bq.L^{-1})	Adult	Child	Infant	H_{inh} (µSv.y^{-1})
1	99 ± 15	1.02 ± 0.15	0.017	0.034	0.019	0.25
2	82 ± 21	0.84 ± 0.22	0.014	0.028	0.016	0.21
3	71 ± 13	0.73 ± 0.13	0.012	0.024	0.014	0.18
4	89 ± 18	0.91 ± 0.19	0.015	0.030	0.017	0.22
5	131 ± 30	1.35 ± 0.30	0.022	0.044	0.026	0.33
Mean	94 ± 23	0.97 ± 0.34	0.016	0.032	0.019	0.24

Table 4 | Radon concentrations and the AED due to ingestion (H_{ing}) and inhalation (H_{inh}) of radon in water samples from the utility water system

	Radon concentration		H_{ing} (mSv.y^{-1})			
S/N	C_{air} (Bq.m^{-3})	C_{water} (Bq.L^{-1})	Adult ×10^{-3}	Child ×10^{-3}	Infant ×10^{-3}	H_{inh} (µSv. y^{-1})
1	51 ± 17	0.52 ± 0.17	8.54	17.08	9.96	0.13
2	100 ± 19	1.03 ± 0.20	16.92	33.84	19.74	0.25
3	45 ± 12	0.46 ± 0.12	7.56	15.12	8.81	0.11
4	39 ± 17	0.40 ± 0.18	6.57	13.14	7.67	0.10
5	23 ± 11	0.24 ± 0.11	3.94	7.88	4.60	0.06
Mean	52 ± 29	0.53 ± 0.30	8.71	17.41	10.15	0.13

Annual effective dose

The risk of radon in water to human health is its direct ingestion by drinking water and the inhalation of radon gas escaping from the water into indoors. The annual effective dose (AED) due to the ingestion of radon in water was determined using the following expression (Tabassum & Mujtaba 2012; Pinti *et al.* 2014; Thabayneh 2015):

$$H_{ing} = C_{water} \times D_{ing} \times L \quad (2)$$

where H_{ing} is the AED in mSv.y^{-1}, C_{water} is the radon concentration in water in Bq.l^{-1}; D_{ing} is the dose conversion coefficient in Sv.Bq^{-1} and L is the annual water intake in L. y^{-1}. The AED was calculated for adults, children and infants. The dose conversion coefficients are given as 1×10^{-8}, 2×10^{-8} and 7×10^{-8} Sv.Bq^{-1} for adults, children and infants, respectively (UNSCEAR 1993). According to Howard & Bartram (2003), the quantity of water required for hydration should be a minimum of 2 L for average adults in average conditions, rising to 4.5 L per day under conditions of raised temperature and/or excessive physical activity, which is typical for Nigeria. They further stated that this figure can be interpreted as applying to all adults and children, given the difficulty in determining whether the ration of adult/child water requirements would remain the same with increasing activity and/or temperature. Therefore, adopting a daily intake of 4.5 L (both adults and children) and the 0.75 L (infants) given by WHO (2004) for 365 days, the H_{ing} for each category of people were estimated and the results are presented in Tables 1–4. The mean AED for adults, children and infants were 0.415, 0.831 and 0.485 mSv.y^{-1}, respectively for boreholes and 0.137, 0.273 and 0.159 mSv.y^{-1}, respectively, for wells. For packaged water, the mean AED were 0.016, 0.032 and 0.019 mSv.y^{-1} for adults, children and infants, respectively and that of utility water were 8.71×10^{-3}, 17.41×10^{-3} and 10.15×10^{-3} mSv.y^{-1}, respectively. Of all the results obtained the mean AED due to ingestion of radon in water from boreholes and wells for adults, children and infants were higher than the recommended reference dose level of 0.1 mSv over one year's consumption of water, which comprises 10% of the intervention exemption level recommended by the ICRP (WHO 2008).

The AED due to inhalation of radon from drinking water was calculated using the expression according to Sujo *et al.* (2004) and used by Thabayneh (2015):

$$H_{inh} = C_{air} \times R \times F \times T \times 9\,\text{nSv.(Bq.h.m}^{-3})^{-1} \quad (3)$$

where H_{inh} is the AED due to inhalation in nSv.y^{-1}, C_{air} is radon concentration in air in Bq.m^{-3}, R is air–water concentration ratio given as 10^{-4} by UNSCEAR (2000). The equilibrium factor between indoor radon and its progeny, F, is given as 0.4 and T is the occupancy factor (assumed to be 7,000 h.y^{-1}) and 9 nSv.(Bq.h.m^{-3})$^{-1}$ is the dose conversion factor (UNSCEAR 2000). The AED due to inhalation ranged from 0.87 to 24.16 μSv.y^{-1} with a mean value of 6.20 ± 5.93 μSv.y^{-1} for boreholes, 0.22–6.25 μSv.y^{-1} with a mean value of 2.04 ± 1.74 μSv.y^{-1} for wells. It varied between 0.18 and 0.33 μSv.y^{-1} with a mean of 0.24 ± 0.06 μSv.y^{-1} for the packaged water and between 0.06 and 0.25 μSv.y^{-1} with a mean value of 0.13 ± 0.07 μSv.y^{-1} for utility water. The results obtained are much less than those due to ingestion. Hence the contribution to dose through inhalation can be ignored.

CONCLUSIONS

^{222}Rn concentration levels in different sources of the water were measured using a portable pulse-counting ionization chamber (alpha spectroscopy), AlphaGUARD (SAPHYMO GmbH). The AED due to ingestion and inhalation of radon in the water samples were determined. Borehole water had the highest value of radon concentration with about 67% higher than the value recommended by the United States Environmental Protection Agency for drinking water. The mean AED of radon concentration in water from boreholes and wells for adults, children and infants were higher than the recommended reference dose level of 0.1 mSv from one year's consumption of water given by the World Health Organization.

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