Measurement of radon levels in water and the associated health hazards in Jazan, Saudi Arabia

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ABSTRACT
Inhalation and ingestion of radon gas constitute grave health hazards. Water of different types and resources is one of the famous radon sources beside soil, building materials, and natural gas. The dissolved radon concentrations in drinking and ground water samples collected from various locations in Jazan area, Saudi Arabia were measured by using sealed cup technique. About 110 water samples were collected from ground and drinking water from 11 different locations. The weighted mean value of the radon activity levels in drinking and ground water were 2.47 ± 0.14 and 2.95 ± 0.22 Bq/L, respectively. Also, the weighted mean of the total annual effective doses from ingestion and inhalation of drinking and ground water were 24.25 ± 1.33 and 28.99 ± 2.12 µSv respectively, which is lower than the safe limit of 0.1 mSv/y. Results reveal that there is no significant public health risk from radon ingested and inhalation of drinking and ground water in the study area.

1. Introduction
Indoors radon gas comes from many sources, such as rock or soil adjacent to house, earth-based building materials, natural gas, and water supplies. Radon is soluble in water, its solubility decreases rapidly with temperature. When a tap or shower is turned on, some of the dissolved radon is released into indoor air. This adds to the radon present from other sources and will give rise to a radiation dose when inhaled (WHO, 2011). It has been estimated that 10 Bq/L of radon in water will, on average, increase the indoor radon concentration by 1 Bq/m³ (UNSCEAR, 1993).

Radon in water can enter the human body in two different ways. Firstly, radon in drinking water can enter the human body through the gastrointestinal tract to deliver the ingestion radiation dose. Secondly, radon can escape from household water and contributing to the radon concentration in air, which can then enter the human body through the respiratory tract to deliver the inhalation radiation dose (Shilpa, Anandaram, & Mohankumari, 2017). Both mechanisms constitute potential health hazards.

Elevated concentration of radon in drinking water causes stomach cancer (Nasir & Shah, 2012). The cancer risk arising from ingested radon is derived from calculations of the dose absorbed by the tissues has estimated that about 30% of the activity concentration of radon in the stomach was integrated in the walls of the stomach (NRC, 1999).

The World Health Organization (WHO, World Health Organization, 2011) estimated that 1–7% of all lung cancer deaths are due to high levels of radon in water and that 10–15% of total indoor radon may be attributed directly to out gassing from tap water (Collman, Loomis, & Sandler, 1991). According to EPA (United States Environmental Protection Agency) regulations, the maximum contamination level for radon in drinking water is 11 Bq/L (USEPA, 1999).

Long period measurements based on using passive dosimeters is advantage because it can integrate the results over long exposure period. For this reason sealed cup technique fitted with CR-39 as a SSNTD is used for monitoring radon level in ground and drinking water from different locations in Jazan province. Based on ICRP and UNSCEAR recommendation, the annual effective dose from ingestion and inhalation of radon can then be estimated.

2. Experimental set up and theoretical approach

2.1. Study area
Jazan region (also known as Gizan) is located in the south west of the Kingdom of Saudi Arabia between the longitudes (42–43) east and latitudes (16–17) in the north and it is sub-divided into 14 governorates. It stretches 300 km along the southern Red Sea coast, just north of Yemen. It covers an area of 11,671 km² and has a population of 1.6 million at the last census. There exists a great variety on dwelling styles, depending on the social and economic level of the populations in the different towns in Jazan.
In this work, 11 different locations covering Jazan region were targeted to measure the radon activity levels in drinking water and groundwater that the population use. The main source of drinking water is the desalination and bottled water, whereas the ground water in many town is used in household as a tap water or in agriculture. A total of 110 water samples were collected from different locations in Jazan area (10 samples/location).

2.2. Samples types and preparation

The main water types used in Jazan city are well, tap, and desalinated water. Water wells are processed in treatment and purification station and supplied to the populations through pipelines to be used as a tap water which is not suitable for drinking. In all studied sites, drinking water is provided from desalinated water collected from many local stores. Groundwater are collected directly from the wells located in houses or in farms. Also, Zamzam holy water is measured which is imported from Makkah and used directly for drinking as an Islamic ritual. Figure 1 shows the distribution of area under study and samples location.

Five samples from each type (0.33 L) is collected and packed in a polyethylene pack to be ready for measurements inside the sealed cup. The advantage of polyethylene is to prevent the evaporation during the long measurement period and then no accumulation of water drops on the detector surface without affecting the penetration of radon to the detection volume of the sealed cup (Sharaf & Abo-Elmagd, 2005).

2.3. Sealed cup technique

A volume of 0.33 L of water was placed in a sealed glass cup of $1.33 \times 10^{-4} \text{ m}^3$ volume with $9.5 \times 10^{-3} \text{ m}^2$ of sample surface area. The cups were fitted with CR-39 detector (TASTRAK) fixed on the top inside of the cup so that the sensitive part of the detector was facing emanating radon gas from the sample. In this configuration, CR-39 detector could record the alpha particles resulting from the decay of radon and its daughters in the whole air volume above the sample. At the end of exposure period (90 days), CR-39 detectors are collected, etched together under their optimum conditions of 6.25 N NaOH at 70° for 6 h; and counted under optical microscope of 400× magnification power.

The sealed cup is calibrated; with the same conditions, of sample to air volume for exhalation rate measurements prior to its use in this work. In the present work, the water volume ($V_w$) should be equal to, or less than 1/3 the cup volume ($V_o$), that is, $V_w \leq 1/3 V_o$ to reduce the effect of back diffusion of radon into the sample as detailed in the work of Abo-Elmagd and Manal Daif, (2010) in the calibration process.

Figure 1. Map of Jazan showing the distribution of sampling sites.
2.4. Radon activity concentration in water

The concentration of radon emanated from the water sample is allowed to build up with time \( t(h) \). The buildup of radon concentration \( C(t) \) verifies the following equation (Al-Jarallah, Abu-Jarad, & Rehman, 2001):

\[
C(t) = C_0(1 - e^{-\lambda t})
\]  

(1)

where \( C_0 \) (Bqm \(^{-3}\)) is the equilibrium radon concentration and \( \lambda \) \((7.55 \times 10^{-3} \text{ h}^{-1})\) is the radon decay constant.

The integration of Equation (1) with time \((t)\) gives the integrated radon concentration \( C_i \) (Bqm \(^{-3}\)d) which simulate the detector accumulation of radon concentration \( C(t) \) over the exposure period, where:

\[
C_i = C_0 \left( t - \frac{1 - e^{-\lambda t}}{\lambda} \right) = C_0 \ t_{\text{eff}}
\]  

(2)

Where \( t_{\text{eff}} \) is the effective exposure time.

Also, \( C_i \) (Bqm \(^{-3}\) d) equals the ratio of the measured track density \( D \) (Tcm \(^{-2}\)) and the detector calibration factor \( K \) (Tcm \(^{-2}\)/Bqm \(^{-3}\) d) (i.e., \( C_i = \frac{D}{K} \)); therefore, the equilibrium radon concentration \( C_0 \) can be written as:

\[
C_0 = \frac{D}{K_{\text{eff}}}
\]  

(3)

The mass exhalation rates \( E_M \) (Bq Kg \(^{-1}\) h \(^{-1}\)) and the radon concentration in water \( C_w \) (Bq/l) is given by (Abo-Elmagd, Soliman, & Daif, 2009; Singh, Sengupta, & Prasad, 1999):

\[
E_M = \frac{C_0 V_o \lambda}{M}
\]  

(4)

\[
C_w = \frac{E_M}{\lambda} = \frac{C_0 V_o}{M} = \frac{C_0 V_o}{V_w}
\]  

(5)

where \( V_o \) (m \(^3\)) is the volume of the sealed cup, \( M \) (Kg) is the mass of water which is equal to \( \rho V_w \) where \( V_w \) is the water volume in liter and \( \rho \) is the water density (equal to 1 kg/l).

2.5. Correction for back diffusion

Radon concentration inside the sealed cup increases with time from zero to its equilibrium value. After reaching equilibrium, radon has significant probability of diffusing back into the sample. This back diffusion results in the reduction of the radon concentration in the vessel and consequently causes an underestimate of the measured mass exhalation rate (Chao, Tung, Chan, & John, 1997; Kumar & Chauhan, 2013).

Back diffusion effect can be easily taken into account if the used decay constant of radon \( \lambda \) in Equations 4 and 5 is replaced by \( \lambda^* = \lambda + \lambda_b \), where \( \lambda_b \) is the decay constant correcting for the first order of removal of radon by back diffusion (Konstantin Kovler, 2006). From the work of Mee, Kang, and Hyun Moon (2005) \( \lambda_b \) is equal to:

\[
\lambda_b = \frac{\lambda V_o}{V_w}
\]  

(6)

In this case Equations 4 and 5 can be rewrite as:

\[
E_M = \frac{C_0 V_o \lambda^*}{M}
\]  

(7)

\[
C_w = \frac{E_M}{\lambda^*} = \frac{C_0 V_o}{M} = \frac{C_0 V_o}{V_w} \left( \frac{\lambda^*}{\lambda} \right)
\]  

(8)

Where, \( \lambda^* \) is the corrected radon decay constant which simulates the removal of radon inside the sealed cup due to its decay (\( \lambda \)) and its back diffusion into the water (\( \lambda_b \)); and we can safely neglect leakage from the sealed cup (Abo-Elmagd, 2014).

2.6. Ingestion and inhalation doses of radon

The annual effective dose for ingestion \( \text{AED}_{\text{ing}} \) (\( \mu \text{Sv/y} \)) was calculated using the following equation (UNSCEAR, 1993):

\[
\text{AED}_{\text{ing}} = C_W \times D_W \times DCF \times T = 7.3 \times C_W \ (\mu \text{Sv/y})
\]  

(9)

where, \( C_W \) (Bq/l) is the mean radon activity concentration in water, \( D_W \) is the daily water ingesting (2 L/day), DCF is the ingesting dose conversion factor of radon and its progeny \((10^{-8} \text{ Sv/Bq})\), and \( T \) is equal to \(365 \text{ day/y} \) (UNSCEAR, 2000; WHO, World Health Organization, 2004).

The annual effective dose of inhalation \( \text{AED}_{\text{inh}} \) (\( \mu \text{Sv/y} \)) was calculated from the following relation (UNSCEAR, 1993):

\[
\text{AED}_{\text{inh}} = C_W \times R \times D \times T \times F = 2.52 \times C_W \ (\mu \text{Sv/y})
\]  

(10)

where, \( C_W \) in Bq/l is the mean radon activity concentration in water, \( R \) is the ratio of radon in air to radon in water which is equal to \(10^{-4} \) or \(10^{-1} \text{ Bq/m}^3/\text{Bq l}^{-1} \) which mean that a concentration of 10 Bq/l in water gives 1 Bqm \(^{-3}\) in air, \( D \) is the dose conversion factor (9 nSv/h per Bq/m \(^3\)), \( F \) is the indoor equilibrium factor between radon and its progeny \((0.4) \) and \( T \) is the indoor time \((7000 \text{ hy}^{-1}) \) (ICRP, 1993; UNSCEAR, 2000).

The total annual effective dose \( \text{AED}_T \) is the sum of \( \text{AED}_{\text{ing}} \) and \( \text{AED}_{\text{inh}} \).

3. Results and discussion

The radon activity levels and the annual effective doses (ingestion and inhalation) are listed in (Table 1) for both drinking and ground waters. For drinking water, the maximum radon level is found in samples from Al-Tahriyah area \((3.82 \pm 0.26 \text{ Bq/l})\) and the minimum in samples from Al-Hijfar area \((1.65 \pm 0.10 \text{ Bq/l})\), the range of the calculated annual effective dose is from 16.3 to 37.5 \( \mu \text{Sv} \) with 21.1 \( \pm \) 5.8 \( \mu \text{Sv} \) average value. For ground water, the radon level ranged from 1.74 \( \pm \) 0.17 Bq/l (Al-Muwasem)
Table 1. Radon activity levels and annual effective doses for drinking and ground water used in different locations in Jazan area.

| Location  | Drinking water | Ground water |
|-----------|----------------|--------------|
|           | $C_{w}$ (Bq/l) | $AED_{ing}$ µSv/y | $AED_{inh}$ µSv/y | $AED_{T}$ µSv/y | $C_{w}$ (Bq/l) | $AED_{ing}$ µSv/y | $AED_{inh}$ µSv/y | $AED_{T}$ µSv/y |
| S1 Al-Muwaseem | 1.74 ± 0.18 | 12.7 ± 1.3 | 4.4 ± 0.5 | 17.1 ± 1.8 | 1.74 ± 0.17 | 12.7 ± 1.1 | 4.4 ± 0.3 | 17.1 ± 1.5 |
| S2 Al-Tuwai | 1.97 ± 0.11 | 14.4 ± 0.8 | 5.0 ± 0.3 | 19.4 ± 1.0 | 2.40 ± 0.30 | 17.5 ± 2.2 | 6.1 ± 0.7 | 23.6 ± 2.9 |
| S3 Al-Hijfar | 1.65 ± 0.10 | 12.1 ± 0.7 | 4.2 ± 0.3 | 16.3 ± 1.0 | 1.82 ± 0.15 | 13.3 ± 1.1 | 4.6 ± 0.4 | 17.8 ± 1.5 |
| S4 Al-Juradyah | 1.86 ± 0.13 | 13.6 ± 0.9 | 4.7 ± 0.3 | 18.2 ± 1.2 | 2.65 ± 0.16 | 19.3 ± 1.2 | 6.7 ± 0.4 | 26.0 ± 1.6 |
| S5 Al-Tahrir | 3.82 ± 0.26 | 27.9 ± 1.9 | 9.6 ± 0.7 | 37.5 ± 2.5 | 2.12 ± 0.21 | 15.5 ± 1.5 | 5.3 ± 0.5 | 20.8 ± 2.0 |
| S6 Al-Harb | 2.14 ± 0.16 | 15.6 ± 1.2 | 5.4 ± 0.4 | 21.0 ± 1.6 | 4.32 ± 0.34 | 31.5 ± 2.5 | 10.9 ± 0.9 | 42.4 ± 3.3 |
| S7 Al-Humra | 1.96 ± 0.14 | 14.3 ± 1.0 | 4.9 ± 0.3 | 19.2 ± 1.3 | 2.00 ± 0.15 | 14.6 ± 1.1 | 5.0 ± 0.4 | 19.7 ± 1.5 |
| S8 Al-Quff | 1.91 ± 0.10 | 14.0 ± 0.7 | 4.8 ± 0.2 | 18.8 ± 0.9 | 3.11 ± 0.51 | 22.7 ± 3.8 | 7.8 ± 1.3 | 30.5 ± 5.0 |
| S9 Al-Shawafrah | 2.11 ± 0.15 | 15.4 ± 1.1 | 5.3 ± 0.4 | 20.8 ± 1.5 | 3.09 ± 0.18 | 22.5 ± 1.3 | 7.8 ± 0.4 | 30.3 ± 1.7 |
| S10 Al-Allah | 2.11 ± 0.15 | 15.4 ± 1.1 | 5.3 ± 0.4 | 20.7 ± 1.4 | 2.56 ± 0.15 | 18.7 ± 1.1 | 6.5 ± 0.4 | 25.2 ± 1.4 |
| S11 Al-Khadrah | 2.36 ± 0.20 | 17.2 ± 1.5 | 6.0 ± 0.5 | 23.2 ± 2.0 | 2.62 ± 0.21 | 19.1 ± 1.6 | 6.6 ± 0.5 | 25.7 ± 2.1 |
| **Range** | 1.65–3.82 | 12.1–27.9 | 4.2–9.6 | 16.3–37.5 | 1.74–4.32 | 12.7–31.5 | 4.4–10.9 | 17.1–42.4 |
| **Average SD** | 2.15 ± 0.59 | 15.7 ± 4.3 | 5.4 ± 1.5 | 21.1 ± 5.8 | 2.58 ± 0.74 | 18.9 ± 5.4 | 6.5 ± 1.9 | 25.4 ± 7.2 |
| **Weighted mean** | 2.0 ± 0.1 | 14.4 ± 0.8 | 5.0 ± 0.3 | 19.4 ± 1.1 | 2.4 ± 0.2 | 17.2 ± 1.3 | 6.0 ± 0.4 | 23.2 ± 1.7 |
to 4.32 ± 0.34 Bq/l (Al-Harub); with average total annual effective doses equal to 25.4 ± 7.2 µSv.

For Zamzam water, the average radon activities level corrected for back diffusion is 1.9 ± 0.15 Bq/l, which is lower than the average value for both drinking and ground water. The ingestion dose from Zamzam intake is about 0.019 µSv/l.

The uncertainties in the measured parameters is ranged from 5 to 10% with 7% average uncertainty in drinking water and ranged from 6 to 16% with 9% average uncertainty in ground water. These different uncertainties make the use of weighted mean more suitable than the average value because the weighted mean is calculated by giving values in the data set more influence according to some attributes of the data such as their uncertainties. The weighted mean as listed in (Table 1) is lower than the average value by about 8%, this indicates that measured values with lower uncertainty have more weight.

The results are corrected for back diffusion effect and summarized in (Table 2). The back diffusion effect depends mainly on the volume of the used water and the sealed cup. To reduce the uncertainty from this effect we used the same volume for all water types and using identical cups. Consequently, the effect of back diffusion was presumably the same in all measurements. However, the correction for back diffusion gave extra 25% for all measurement values.

The radon activities levels in the studied samples did not exceed the maximum level of contamination of 11.11 Bq/l proposed by the US Environmental Protection Agency (USEPA (United States environmental protection agency), 1999).

The total average annual effective dose from radon due to intake of drinking water (24.25 ± 1.33 µSv) is less than the recommended maximum value of 100 µSv y⁻¹ suggested by WHO and EU council (European Union Commission, 2001; WHO, World Health Organization, 2004). But the average dose from ingestion (18.03 ± 0.99 µSv) is higher than the populated-weighted average annual effective dose from ingestion (10 µSv) proposed by (UNSCEAR, 1993). This rate of 10 µSv/y is based on the assumption that the representative population consists of 5% infants, 30% children and 65% adults. The deviation is not annoying because it come from the low value of the annual water intake proposed by (UNSCEAR, 1993) for different ages. Also UNSCEAR proposed that the annual effective doses to children are doubled for ingestion due to the double annual water intake.

Further, the dose contribution due to inhalation and ingestion leads to lungs and stomach dose which is calculated by multiplying the inhalation and ingestion doses by the tissue weighting factor for lung and stomach; respectively. According to the tissue weighting factor of 0.12 (ICRP recommendations of the ICRP, 1991), the average lung and stomach annual doses are 0.6 µSv and 1.7 µSv; respectively.

The obtained radon activity levels in the present study are compared with the literature findings as listed in Table 3 for drinking and ground waters.

### Table 2. Summary of the measured parameters after correction for back diffusion effect.

| Water types | Drinking water | Ground water |
|-------------|----------------|--------------|
| Parameters | Range | Av. ± SD | Weighted mean | Range | Av. ± SD | Weighted mean |
| C<sub>0</sub> (Bq/l) | 2.06–4.78 | 2.69 ± 0.78 | 2.47 ± 0.14 | 2.18–5.40 | 3.23 ± 0.92 | 2.95 ± 0.22 |
| AED<sub>inh</sub> (µSv/y) | 15.07–34.89 | 19.61 ± 5.38 | 18.03 ± 0.99 | 15.91–39.40 | 23.58 ± 6.72 | 21.55 ± 1.58 |
| AED<sub>ing</sub> (µSv/y) | 5.20–12.04 | 6.77 ± 1.86 | 6.22 ± 0.34 | 5.49–13.60 | 8.14 ± 2.32 | 7.44 ± 0.54 |
| AED<sub>t</sub> (µSv/y) | 20.27–46.93 | 26.38 ± 7.24 | 24.25 ± 1.33 | 21.41–53.00 | 31.72 ± 9.04 | 28.99 ± 2.12 |

### Table 3. Comparison of present study with reported values from other countries.

| Water type | Radon activity (Bq/l) | References | Country |
|------------|-----------------------|------------|---------|
| Drinking water Open well | 0.27–5.4 0.24–9.82 | Shilpa et al., 2017 | India |
| Drinking water Ground water | 0.20–1.23 (0.6) 0.42–0.89 (0.64) | Thabayneh, 2015 | Palestine |
| Groundwater | 1.02–7.26 | Amin, 2014 | Libya |
| Drinking water Ground water | 0.333–0.903 (0.662) 0.670–1.45 (1.21) | Nasir & Shah, 2012 | Pakistan |
| Drinking water Ground water | 0.39–0.97 (0.95–36.00) | Marques, Dos Santos, & Geraldo, 2004 | Brazil |
| Ground water | 0.1–5 (1.4) | Sarrou & Pashalidis, 2003 | Cyprus |
| Drinking water Ground water | 1.65–3.82 (2.0) 0.74–4.32 (2.4) | Present work | Saudi Arabia |
Disclosure statement

No potential conflict of interest was reported by the authors.

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