Lifetime cancer risks of radon and heavy metals levels in groundwater wells around limestone quarry in Ogun State, Nigeria

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ABSTRACT

Background: Radon (^{222}Rn) and carcinogenic metals are major contaminants in groundwater wells. This research aimed on evaluating the possible lifetime cancer linked with the presence of radon and carcinogenic metals in groundwater wells situated close to a limestone quarry in Ewekoro, Ogun State, Nigeria. ***Materials and Methods:*** Twenty (20) groundwater wells near a limestone quarry were sampled for water sample collection. Radon levels in water were assessed utilizing CR-39 detectors, whereas concentrations of carcinogenic metals were analyzed via atomic absorption spectrometry. The resultant data facilitated the estimation of potential lifetime cancer risks for adults exposed to water from these wells. ***Results:*** The measured radon concentrations varied between 2.16±1.7 and 11.88±1.6 Bql^{-1}, with an average of 6.67 ± 2.15 Bql^{-1} across the 20 samples. A significant portion, 81%, of the samples exhibited radon concentrations below the permissible limit of 11.1 Bql^{-1} fixed by the United States Environmental Protection Agency (USEPA). The annual effective dose (AED) attributable to inhaled and ingested radon ranged from 5.44±4.3 to 29.94±4.0 µSvy^{-1} and 15.77±13.9 to 86.72±11.7 µSvy^{-1}, respectively. The collective AED across all water sample sources varied from 21.21±16.7 to 116.67±15.7 µSvy^{-1}, with average values of 65.434±10.4 µSvy^{-1}. The cumulative incremental lifetime cancer risk related to the ingested carcinogenic metals adults ranged from 9.70 ×10^{-5} to 1.03×10^{-4} with a mean value of 9.72 ×10^{-5}. ***Conclusion:*** Water wells situated closer to the limestone quarry exhibited higher mean radon concentrations, while those farther away from the quarry maintained mean radon concentrations below the acceptable limits provided by the World Health Organization (WHO) and the USEPA.

INTRODUCTION

Most African countries rely heavily on groundwater, constituting about twenty percent of global fresh water supplies (1), serving different purposes such as drinking, domestic, and industrial uses, significantly affecting human life (2, 3). However, groundwater quality, especially in areas near limestone deposits, is severely affected by quarrying activities driven by high demand for cement and construction materials. Quarrying can disrupt active groundwater conduits, leading to blockages, reduced flow velocity, altered pH levels, and increased concentrations of geogenic heavy metals and radon due to the dissolution of bedrock minerals in the aquifer (4). This decline in water quality poses serious health risks to nearby residents (5-7).

Radon (^{222}Rn), a gas found in the decay series of radium-226, is a significant concern. It is an alpha particle emitter with high linear energy transfer, causing greater biological damage compared to beta and gamma radiation. Radon is short lived with half-life of 3.8 days, depositing all its radiation energy in a short period, making it particularly hazardous when inhaled or ingested (8-10). The carcinogenic effects of radon, potentially leading to lung and gastrointestinal cancers, have garnered increasing research attention (11, 12). The World Health Organization (WHO) recommend a standard limit of 100 Bql^{-1} for radon concentration in ingested water, whereas the USEPA suggests a maximum concentration level of 11 Bql^{-1} (13, 14). This research also focuses on carcinogenic heavy metals, and the ones assessed in this study are chromium (Cr), cadmium (Cd), and lead (Pb). Long-term and repeated exposure to elevated levels of ingested lead (Pb), chromium (Cr), and cadmium (Cd) in water has been linked to risk of developing gastrointestinal, prostate, and kidney cancers (12, 15–18).

Binesh and colleagues conducted an evaluation of radon levels in 50 tap water samples from Mashhad, Iran, utilizing the Portable Radon Gas Surveyor

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SILENA. The mean radon concentration was 16.238 ± 9.322 Bql^{-1}, with 70% exceeding the USEPA threshold of 11 Bql^{-1} for radon level in consumable water [9].

Malakootian *et al.* studied radon in 27 drinking water samples near Bam Village, Iran, with RAD7. The average maximum radon concentration measured in drinking water around geological faults was 9.88 Bql^{-1}, below EPA and WHO guidelines [10]. Najam and colleagues used CR-39 detectors to measure radon in Wassit governorate, Iraq, the findings showed that radon concentrations ranging from 0.325±0.02 to 0.563±0.12Bql^{-1}, below the USEPA permissible limit, were observed [19].

Ahmad *et al.* and Ajiboye *et al.* separately did detailed studies in Malaysia and Nigeria, respectively, to assess the health hazards associated with radon levels and heavy metal presence in drinking water. Ahmad et al. noted that groundwater wells exhibited elevated radon concentrations compared to tap water. Ajiboye *et al.* also observed some higher concentrations of radon recorded above the safe limit and elaborated that the water wells in sedimentary basements contain more dissolved heavy metals than igneous [20, 21].

This current study explores the relationship between radon and heavy metals in groundwater wells, especially in areas near limestone quarries, where continuous dissolution of heavy metals and radon due to quarrying activities may poses health risks to inhabitants. Notably, there are no extensive researches on measuring concentrations of radon and carcinogenic metals and assessing associated health risks in groundwater around limestone quarries. Hence, the objective of this research is to analyze the levels of radon, lead, chromium, and cadmium within selected groundwater samples, and evaluate the potential cancer risks near Ewekoro limestone quarry in Ogun State, Nigeria.

MATERIALS AND METHODS

Geology of the research area

The research area (figure 1) is at a limestone deposit with active quarry activity. It is situated between latitudes 60°48'N and 60°56'N and between longitudes 30°13'E and 30°38'E. The area is part of the Dahomey basin. Records indicate that the rock units of the study region are situated in the Ewekoro Depression, a low-lying marshy depression in southwest Nigeria that is a part of the Dahomey Basin [22, 23]. The enormous coastal sedimentary basin known as the Dahomey Basin is found on the edge of the Gulf of Guinea. The Paleocene formation is an essential part the geologic formations in the Dahomey Basin. Glauconitic limestone dominates, with trace amounts of shale, marl, and sand dispersed throughout the region [23].

Water sampling

Twenty (20) samples were collected from 20 groundwater wells in January 2023 during the dry season and the peak period of quarry activities at the study area. The wells were positioned at a distance spanning from 500 meters to 900 meters away from the quarry. The samples were collected from the groundwater wells in 250 ml glass vials with a rope on the bottle's neck to draw from the wells. Upon retrieval from the wells, sample containers were promptly sealed with their respective caps to stop the introduction of air bubbles and the release of radon gas. Subsequently, the samples were meticulously labeled, timed and stored in ice-filled containers. This protocol represents the established methodology for the collection and conservation of groundwater samples intended for analytical purposes [24, 25].

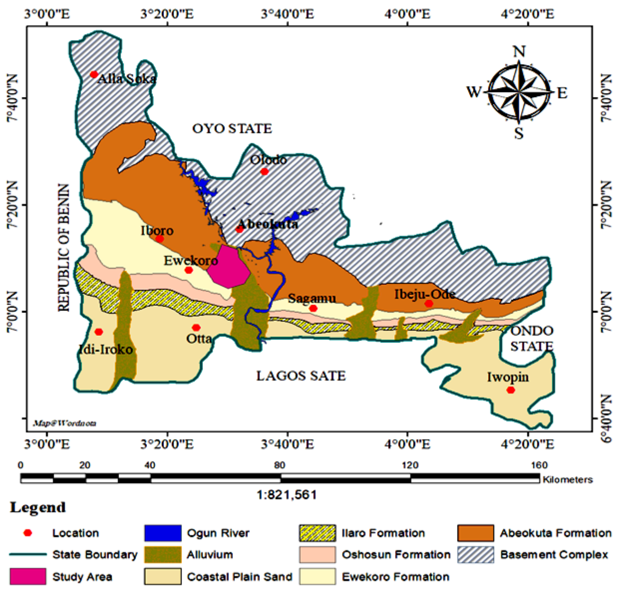

Figure 1. Geologic map and location of research area.

Physio-chemical and atomic absorption spectroscopy (AAS) analysis

The electrical conductivity (EC) and pH were determined using pH/EC meter (Eijkelkamp Agrisearch equipment, model: 18.50.SA portable multimeter, Netherland). The electrode was rinsed with distilled water. The manufacturer's calibration liquid was used for calibration before measurements on the groundwater at an average temperature of 25˚C. The collected samples underwent analysis for three heavy metals (lead, chromium, and cadmium) employing established techniques for water quality assessment. Heavy metal concentrations in the water samples were determined utilizing an atomic absorption spectrophotometer (Shimadzu, Model: AA -7000, Japan). A lamp was utilized as the light source, emitting appropriate wavelengths, within an acetylene flame for measurements. Calibration of the equipment was conducted at its maximum operational sensitivity in agreement with the

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manufacturer's specifications.

Radon activity measurements

Radon measurement was done with CR-39 detectors (Pershore Moulding LTD Co. UK). The thickness of the CR-39 detector is about 500 µm and its area is 1×1 cm^2. The alpha particle interacts with and penetrates the surface of the CR-39 and causes damage along its path, which becomes visible by chemical etching. 250 ml of stored water samples from each well in the study were transferred to the laboratory just 2 hours after collection. During this time, it is expected for the thoron (with a half-life of 57 seconds) to have decayed. The water samples were each poured separately, without the residue (if any), into sealed chambers such that the air space between the water surface and the detector is 10 cm, as shown in figure 2. The setups are then placed in a cool, isolated part of the laboratory for 30 days to attain secular equilibrium. The half-life of radon is about 4 days, so within 30 days, 7 times its half-lives are sufficient for the gas to reach a secular equilibrium between ^{226}Ra and ^{222}Rn, or ^{222}Rn and ^{218}Po [26].

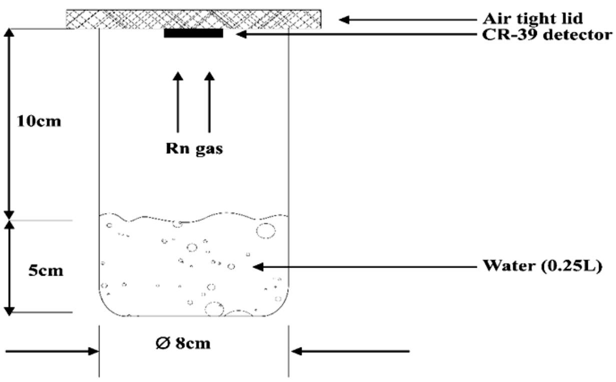

Figure 2. Radon measurement in water sample using sealed-cup technique.

After 30 days of exposure, the CR-39 were etched in a 6M NaOH solution for 4 hours at a constant temperature of 80°C in a water bath. The enlarged alpha tracks on the detectors analyzed under an optical PC-aided microscope system with a magnifying power of 400X, and the alpha track densities were analyzed using ImageJ software semi-automated counting. The radon gas concentrations C_{Rn} (Bq/m3) on the exposed CR-39 were calculated as follows in equation (1) [19].

$$C_{Rn} = \frac{C_\circ}{\rho_\circ} \times \rho \qquad (1)$$

Where; $C_\circ$ is the radon concentration in standardized water sample (Bq/m^3), $\rho_\circ$ is the track density (tracks /cm^2) on exposed CR-39 in standard water and ρ is the track density (tracks/cm^2) on exposed CR-39 in collected water samples.

Estimation of annual effective dose of radon

The pathways through which radon from water can enter the human body include inhalation and ingestion, with the lungs and stomach being the primary organs of concern. The absorbed radiation dose by the stomach through ingestion depends on the drinking of water. In addition, during other human activities such as inhalation just about to drink water, bathing, laundering, dishwashing, etc., there is a possible radiation dose to the lungs. In order to calculate the AED from the inhalation and ingestion of radon found in water, one must take into account the radon concentration in both air and water, along with the duration of exposure, and the conversion dose factors associated with radon ingestion or inhalation. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 2000 model established formulas to estimate AED to the public from radon in equations 2 and 3 [10].

$$E_{inh}\ (\mu Svy^{-1}) = C_w \times R_{a,w} \times E_f \times T \times D_{inh} = 2.52 \times C_w \qquad (2)$$

$$E_{ing}\ (\mu Svy^{-1}) = C_w \times W_r \times D_{ing} = 7.3 \times C_w \qquad (3)$$

Where; E_{inh} is the AED from inhaled radon freed from water into air (µSvy^{-1}), C_w is the radon level in water (Bql^{-1}), $R_{a,w}$ is the ratio of radon in air to radon in water (10^{-4}), and T is the average indoor time of occupants (7000 hy^{-1}). E_f is Equilibrium factor between radon and its progenies (0.4), D_{inh} is the conversion dose factor for radon exposure for adults (9 nSv(Bqhm^{-3})$^{-1}$), E_{ing} is the AED from drinking water containing radon (µSvy^{-1});W_r is the volume rate of annual water consumption = 730 l/y; D_{ing} is the adults' conversion dose factor = 10^{-8} nSv Bq^{-1}.

Carcinogenic health risk assessment

Assessing cancer risks (CR) entails probabilistically determining a person's likelihood of developing cancer throughout their lifetime because of exposure to potentially toxic metals [27, 28].

The United States Environmental Protection Agency (USEPA) models in equations (4) and (5) were employed to compute the carcinogenic risks. Initially, this involved determining the chronic daily intake (CDI) via the ingestion pathway specifically for adults [14].

$$CDI_{ing} = \frac{C_w \times IR \times ED \times EF}{B_w \times AT} \qquad (4)$$

CDI_{ing} is ingested Chronic Daily Intake the exposure dose (mg kg^{-1}day^{-1}), C_w is the mean concentration of the trace elements in water (mg l^{-1}) and IR is the intake rate of drinking water (2 liter day $^{-1}$). ED is the exposure duration (70 years), EF is the exposure frequency to pollutants (365 days/year), BW is the total body weight (70 kg) and AT is the ED×365 for non-carcinogenic risk, (2555).

The carcinogenic risk from drinking groundwater contaminated with potentially toxic metals was calculated using the equation (5) [30].

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$$CR_{ing} = CDI_{ing} \times CSF \quad (5)$$

Where; CR_{ing} is the carcinogenic risk from drinking contaminated groundwater, and CSF is the Cancer Slope Factor (CSF) value (mg kg⁻¹ day⁻¹) for oral intake.

The USEPA model gives CSF for Cr, Cd and Pb to be 0.5, 15 and 0.009 mg/kg/day respectively [31, 32]. The total incremental lifetime cancer risk (ΣILCR) for carcinogenic metals ingested in the groundwater samples for the three metals is given by the equation (6).

$$\sum \text{ILCR} = CR_{ing}^{Cd} + CR_{ing}^{Cr} + CR_{ing}^{Pb} \quad (6)$$

Statistical analysis

The analysis of the results and the generation of the plots were statistically performed using Statistical software, qtGrace 0.2.3 Beta. Statistical significance of p-value of < 00.05 was considered for the measurement of the radon and heavy metals in water.

RESULTS

Radon concentrations

The results of radon concentrations and distributions are described in table 1 and figure 3. Radon concentrations ranged from 2.16±1.7 Bql^{-1} to 11.88±1.6 Bql^{-1}, with a mean value of 6.67 ± 2.15 Bql^{-1}. Table 2 and figure 4 show the mean radon levels in water wells samples collected around the quarry. The highest level of 11.88±1.6 Bql^{-1}was recorded at a well within 500–600 m away from the quarry. The mean radon level in the water wells at varying ranges of distances from the quarry reached its peak (11.88 Bql^{-1}) at the closest distance range. The mean radon values decrease significantly to 4.536 Bql^{-1} (600–700 m), and then increase again to 5.4 Bql^{-1} (700–800 m), and later to 7.98 Bql^{-1} (800–900 m).

Effective radiation dose assessment

The AED of radon in drinking water samples wells around a limestone quarry is presented in table 2. The total AED of all the water sample sources in relation to the reference dose level (RDL). The AED ranged from 5.44±4.3 μSvy^{-1} to 29.94±4.0 μSvy^{-1}, while 15.77±13.9 μSvy^{-1} to 86.72±11.7 μSvy^{-1}, respectively. The cumulative annual effective dose (AED) across all water sample sources ranged from 21.21±16.7 μSvy^{-1} to 116.67±15.7 μSvy^{-1}, with an average of 65.434±10.4 μSvy^{-1}. Analysis revealed that precisely 10% of the total AED exceeded 100 μSvy^{-1}, indicating potential health risks from ingestion and inhalation. Specifically, water samples from groundwater wells T4 and T18, situated at distances of 200–350 m and 650–800 m from the quarry, respectively, exhibited slightly elevated readings surpassing 100 μSvy^{-1}.

Table 1. Radon concentration in water CRn (Bql^{-1}) and annual effective dose (AED) for water samples collected in groundwater wells at the study area.

Samples ID	C_{Rn} (Bql^{-1})	E_{inh} (μSv/y)	E_{ing} (μSv/y)	AED (μSvy^{-1})
T1	8.20±2.5	20.66±6.3	59.86±19	80.52±24.6
T2	5.25±3.2	13.23±8.1	38.33±23.6	51.56±31.4
T3	9.72±2.2	24.49±5.5	70.96±16.1	95.45±21.6
T4	11.88±1.6	29.94±4.0	86.72±11.7	116.67±15.7
T5	8.64±3.5	21.77±8.8	63.07±25.6	84.84±34.4
T6	3.24±1.1	8.17±2.8	23.65±8.0	31.82±10.8
T7	5.4±1.3	13.61±3.3	39.42±9.5	53.02±12.7
T8	2.16±1.9	5.44±4.3	15.77±12.4	21.21±16.8
T9	9.72±2.5	24.49±6.3	70.96±18.3	95.45±24.6
T10	2.16±1.9	5.44±4.8	15.77±13.9	21.21±18.7
T11	3.24±1.4	8.16±3.5	23.65±10.2	31.82±13.8
T12	9.72±2.2	24.49±5.5	70.96±16.1	95.45±21.6
T13	6.48±3.3	16.33±8.3	47.30±24.1	63.63±32.4
T14	5.4±.1.5	13.61±3.8	39.42±10.9	53.03±14.7
T15	2.16±1.7	5.44±4.3	15.77±12.4	21.21±16.7
T16	7.56±3.6	19.05±9.1	55.19±26.3	74.24±35.4
T17	5.65±1.8	14.24±4.5	41.25±13.1	55.48±17.7
T18	10.2±2.2	25.71±5.5	74.46±16.1	100.16±21.6
T19	7.54±2.9	19.01±7.3	55.04±21.2	74.04±28.5
T20	8.95±4.4	22.55±11.1	65.34±32.1	87.89±43.2

Table 2. The average radon concentration at varying distances away from the limestone quarry.

Samples ID	Distance from Quarry (m)	C_{Rn} (Bql^{-1})	E_{inh} (μSvy^{-1})	E_{ing} (μSvy^{-1})	AED (μSvy^{-1})
T1 - T5	500 - 600	8.74	22.11	63.79	85.81
T6 -T10	600 -700	5.25	13.23	38.33	51.56
T11 - T15	700 -800	5.40	13.61	39.42	53.03
T16 -T20	800 - 900	7.98	20.12	58.25	78.36

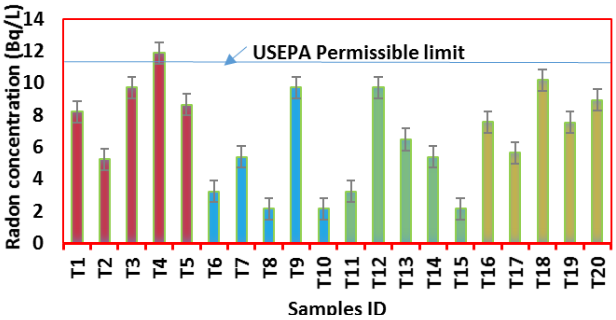

Figure 3. Radon consternation in groundwater wells around limestone quarry, Ewekoro, Ogun State.

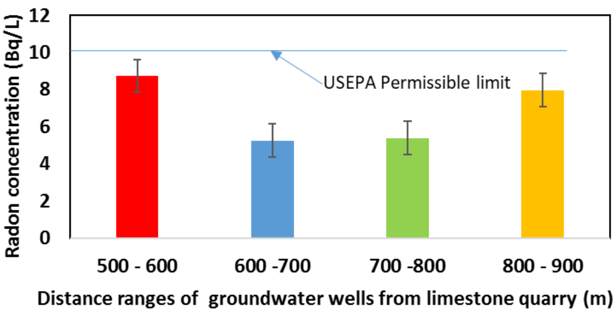

Figure 4. Radon consternations variation with range of distances in groundwater wells from limestone quarry, Ewekoro, Ogun State.

Physicochemical properties and heavy metal concentration

Table 3 gives the range of the metals

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concentrations in mgl^{-1}are as follows: for Cd: 0.003 to 0.008, Pb: 0.01 to 0.065, and Cr: 0.002 to 0.008. The decreasing order of the mean concentration of the metals are Pb (0.01655 mgl^{-1}) > Cr (0.00875 mgl^{-1}) > Cd (0.0056 mgl^{-1}). The electrical conductivity of the sampled water had a range of 600 to 2800 μScm^{-1} with an average value of 1266.4 μScm^{-1}.

Carcinogenic health risk

The estimated cancer risk exposure from ingesting the heavy metals among the residents of the research region is presented in table 3. According to the findings, the computed mean incremental lifetime cancer risk (ILCR) values for ingesting Pb, Cr, and Cd were 6.27 × 10^{-7}, 5.42 × 10^{-5}, and 4.24 × 10^{-5} respectively. The total incremental lifetime cancer risk of the carcinogen in all the water samples collected from the groundwater wells ranged from 2.07× 10^{-5} to 2.01× 10^{-5} with mean value of 9.72× 10^{-5}.

Table 3. Physiochemical parameters, concentration and calculated incremental lifetime cancer risk (ILCR) values of Cr, Pb and Cd in drinking water samples collected from Ewekoro, Ogun state.

Number of samples (N = 20)	EC (µS/cm)	pH	Concentration (mgl^{-1}) Cr	Pb	Cd	Calculated cancer risks Cr	Pb	Cd	∑ILCR
Minimum	600	7.2	0.003	0.01	0.002	5.20E-07	8.64E-08	7.80E-06	2.07E-05
Maximum	2800	8.5	0.008	0.065	0.008	7.11E-06	2.16E-06	1.07E-04	2.01E-04
Average	1266.4	7.5	0.0166	0.00875	0.0056	5.42E-05	6.27E-07	4.24E-05	9.72E-05

DISCUSSION

Radon concentration levels were measured in 18 well water samples collected near a limestone quarry in Ewekoro. Most samples showed radon concentrations below 11 Bql^{-1}. However, two samples exceeded the recommended upper permissible limit of 11.1 Bql^{-1} set by the USEPA [14]. According to WHO, immediate remediation is advised for radon concentrations in drinking water exceeding 100 Bql^{-1} [13]. The UNSCEAR suggests a range of 4 to 40 Bql^{-1} for ^{222}Rn activity levels in water for human consumption [22]. Despite these recommendations, Nigeria has yet to establish guidelines and regulations concerning radon concentration in drinking water. There are no significant differences in radon levels compared to previous results recorded in other parts of the country; the results obtained are consistent, having relative average values greater than 6 Bql^{-1} [33, 34]. The radon level in groundwater is subjected to several factors such as lithology, fractures and fissures, permeability of the bedrock formation, groundwater flow, depth of the water table, seasonal variations, and human activities like quarrying and mining [35]. In the study conducted by Khan *et al.* on wells water in the limestone region of Karak, Pakistan, the result showed that the highest average ^{222}Rn concentration was 16.5 Bql^{-1}. The research indicated that the mean radon levels observed in limestone were greater compared to those found in gypsum. [6]. A similar study conducted in St. Catherine, Jamaica, by Smith *et al.* shown that the radon concentration (at a mean of value of 18 Bql^{-1} ± 2 Bql^{-1}) in drinking water samples from well water was higher than the USEPA permissible limit [36]. The separate studies carried out by Malakootian *et al.* and Dosunmu *et al*. agreed that the lithograph and the geology of the bedrock of the groundwater wells could influence the radon concentration. The radon concentration in water will become excessively high in areas with geological fractures and active faults [10, 37]. In this study, the aquifer is composed of limestone, which generally has low traces of uranium and radium. The high radon concentration recorded at wells close to the quarry was primarily influenced by mining activity, which further affects the movement of groundwater through the pores of limestone aquifers and can affect the radon concentration. Faster groundwater flow may reduce the time for radon decay, resulting in higher radon levels in the water [6, 36]. Then, as we move far away from the limestone aquifer, the geological composition of the area becomes more granitic, which plays a significant role in elevated radon concentrations in groundwater. The mean concentrations of the carcinogenic metals (Cr, Pb, and Cd) in the samples showed that these values are lower than the WHO permissible limits for Pb and Cr in drinking water, which are 0.05 and 0.5 mgl^{-1} respectively, except for cadmium, which was found to be higher than the acceptable limit of 0.01 mgl^{-1} [13]. The slightly high concentration of Cd is attributed to the geological composition of the bedrock. Quarry activity is a major factor that causes the dissolution of the minerals in the bedrock, leading to the contamination of water and significant health risks when consumed. The electrical conductivity (EC) also revealed the extent of weathering of the bedrock and the amount of dissolved minerals in the groundwater. The value of EC obtained in this study exceeded WHO standards of 400 μScm^{-1}, which indicated that the water in the study area is significantly ionized due to the high level of weathering in the bedrock of the groundwater [13]. In this study, the assessment of cancer risk associated with consuming carcinogenic metals in drinking water indicated that both the incremental lifetime cancer risk (ILCR) and the cumulative lifetime cancer risk (∑ILCR) remained within the acceptable threshold (≤10^{-4}) for cancer incidence in the adult population [38, 39].

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CONCLUSION

This study presents findings on radon and selected carcinogenic metal concentrations in groundwater wells surrounding a limestone quarry in Ewekoro, Ogun State, Nigeria. Results indicate that radon levels in most samples were within safe limits, except for one sample slightly above the USEPA advised threshold. The total annual effective dose was below the WHO safe limit. The total incremental lifetime cancer risk from lead, cadmium, and chromium was within the USEPA acceptable range. Therefore, quarry activities are expected to affect groundwater quality, potentially leading to increased radon and carcinogenic metal concentrations due to water mixing and mineral dissolution.

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