



Assessment of ambient gamma radiation dose and annual effective dose associated with radon in drinking water from gold and lead mining area of Moro, North-Central Nigeria

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Abstract

This work investigates the activity concentration of ^{238}U , the ambient gamma absorbed dose rates (DR), and the ^{222}Rn concentration levels in ground and surface water samples from gold and lead mining area of Moro, Kwara State, Nigeria. In situ measurements of activity concentration of ^{238}U and DR were carried out using Super SPEC RS-125 gamma spectrometer and ^{222}Rn activity concentration in ground and surface waters were analysed using a calibrated RAD7 big bottle system. The results show that the risk of radiation exposure at the mining area is high and the public are not safe from exposure to ionizing radiation.

Keywords Cancer · Radioactivity · Radon · Annual effective dose

Introduction

Human exposure to elevated level of ionizing radiation is very harmful as it can cause cancer and other radiation induced sicknesses [1–3]. Radiation in the environment can come from terrestrial and cosmic and/or the anthropogenic sources (human activities such as mining) [4, 5]. ^{222}Rn an isotope of ^{226}Ra has been one of the major causes of cancer in the world. It is a colourless, odourless and tasteless noble gas with half-life of 3.8 days [6]. This short half-life makes it one of the rarest radionuclides compared to uranium and thorium (mother radionuclides) with very long half-lives (several billions of years). Despite its rare nature, it will continue to be present in the future as it is continually generated from its mother radionuclides (uranium) [7, 8]. Unlike other radionuclides, radon is considered as one of the major radionuclide that poses serious health risk as it is easily inhaled and soluble in underground waters, though sparingly insoluble in surface water. Its level had been known to differ from location to location due to differences in geology

and human activities such as mining, milling, air effluence, industrial solid and liquid wastes, etc. [9].

Epidemiological studies have shown a strong correlation between breathing high concentrations of radon and lung cancer as it is a contaminant that affects both the indoor air and water in the environment. ^{222}Rn is one of the major contaminants in surface and ground water resources in mining areas due to large volume of radioactivity bearing excavated waste that remains after minerals extraction from their ores in mining site [3, 9, 10]. The quantification of radon in water which is highly needed for domestic purpose is very important from the public world perspective. Owing to the importance of water, human beings do anything possible to get water from several sources which includes water bodies (rivers, streams, etc.), rain, and underground (well, bore-holes etc.) [2, 11]. It was observed that cancers caused by chemical carcinogens are majorly obtained from drinking water [12–14].

Since water is often gotten from natural sources, which includes wells, rivers, etc. significant amount of radionuclides: ^{40}K , ^{226}Ra , ^{232}Th , ^{238}U , ^{222}Rn , have over time been found traceable in water. ^{238}U available in the soil slowly disintegrates to other compounds such as radium, which over time eventually becomes radon [15]. A fraction of the ^{222}Rn stays at the sub-surface and penetrates the groundwater, while the others migrates to the soil surface and escape into the air [16]. This radon composed in either surface

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or ground waters may find their way into the human body through inhalation (during degassing) or ingestion (drinking of the water). During either process (ingestion or inhalation), this radon disintegrates to its daughter nuclides giving out alpha particles which irradiates the cells of either the lungs or the stomach and causes it to mutate and in an end result induces cancer [17].

Owing to the previous knowledge of elevated levels primordial radionuclides in other mining areas within Nigeria [9, 18], it is therefore necessary to assess the ambient gamma DR, ^{238}U and ^{222}Rn radioactivity level in waters from the gold and lead mining areas in Moro Local Government Area of Kwara State. This study will help to ascertain the radiological safety in utilizing the waters in this area since no similar work had been carried out to the best of our knowledge.

Materials and methods

Study area

Moro Local government area is located in Kwara State, Nigeria, West Africa, Africa, within latitude 8.8449° or $8^\circ 50' 41.7''$ North and Longitude 4.5567° or $4^\circ 33' 24.2''$ East as presented in Fig. 1 [3, 14, 19–23]. It has elevation of 251 m (823 feet) and a landmass of 3272 km^2 . In 2006 population census, it had a total population of 108,792 people. Moro Local government area is popularly known for the mining of gold and lead ores. Kwara state is known to be underlain by basement complex rocks represented by phyllites, gneisses, granodiorites, granites, schists and pegmatites. Gold mineralization is mostly found to be linked with gneisses and schists in this part of the country [19]. Sandstone

beds at the contact between the Cretaceous sediments of Nupe Basin and the Basement Complex around Bishewa and Gidan Sani in Moro are known to host gold. Evaluation of this resource in panned concentrate of soils and stream sediments around Bishewa, Gidan Sani, Koro and River Wara (surface water) gave between 4.1 and 8.3 g/t, however assay of most of the gneisses found proximal to these areas range between 1.1 and 4.5 g/t. New locations found are Ndanaku, Mari, Oputa, Lokomosi, Tunga Bichi, Gbajubo, Giloadi and Birnin Ruwa in Edu and Ifelodun LGA. Preliminary evaluation and assessment of the resource in Ndanaku and Birnin Ruwa gave gold values averaging 5 g/t. This suggests that the area of gold mineralization is extensive.

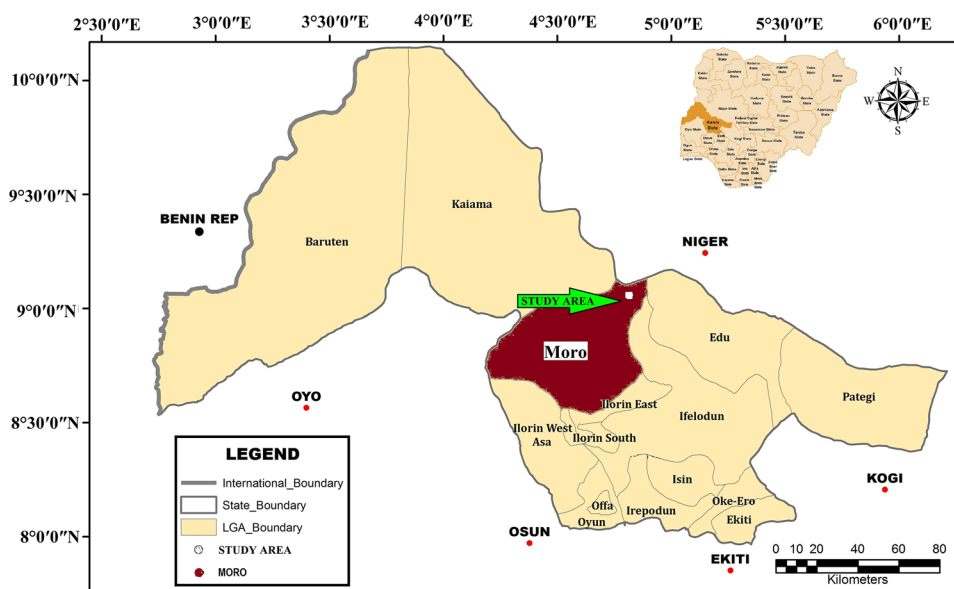
Field survey

In situ measurements of activity concentration of ^{238}U and the gamma radiation dose exposures at the gold and lead-mining fields in Moro LGA of Kwara State, Nigeria was carried out using Super SPEC RS-125 gamma spectrometer. The Super SPEC RS-125 gamma spectrometer has a large 2.0×2.0 NaI crystal that offers superior integrated design with big detector, easy to use and good sensitivity. The in situ measurements were carried out at about 1 m above the earth [10]. The measurements are presented in our data article [19].

Details about the Super SPEC RS-125 gamma spectrometry methods can be found in earlier works where the device was used [10, 19, 21].

The indoor and outdoor annual effective doses for the general public was estimated using the equation below [1, 24]:

Fig. 1 Map of Kwara State, displaying the study area (Moro LGA)



$$\text{AED (mSv/y)} = \text{DR} \times \text{T} \times \text{OF} \times \text{C} \quad (1)$$

where DR is the absorbed dose rate, T is time in hour for 365 days ($T=8760$ h), OF is the occupancy factor (0.2 for outdoor exposure) and C is the conversion coefficient. According to [1] report, the conversion coefficient is 0.7 SvGy^{-1} for dose in air to effective dose received by adults.

Collection and preparation of water samples

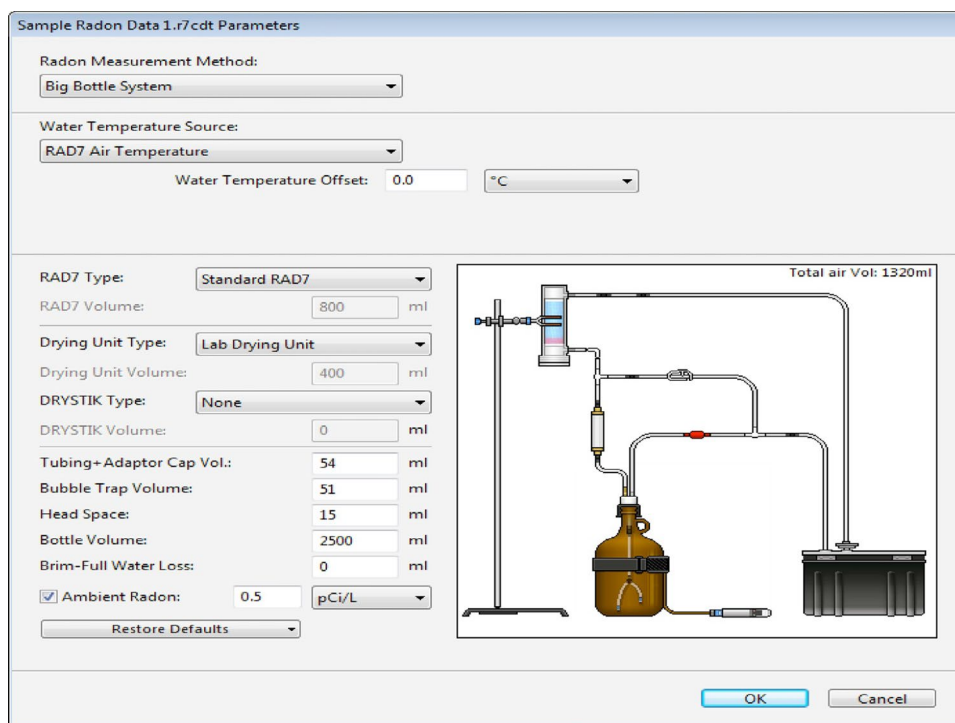
Forty-eight (48) water samples (24 surface and 24 well water samples) were obtained from gold and lead mining site in Moro LGA, Kwara State, Nigeria. Each water sample was used for the analysis. The 48 samples were collected very early in the morning, to avoid certain properties of the water from getting altered by human activities before the time of collection. The samples were collected in a 1.5 l plastic bottle, as large water samples of up to 2.5 l may be sampled using RAD7 big bottle system [25]. The samples were obtained directly from the surface and ground waters into a clean 1.5 l plastic bottle. Prior to use, the bottles were washed thoroughly with distilled water to avoid or minimize contamination or absorption of the radon present in the samples. The plastic bottles were dipped directly into the fetcher containing the water and covered right inside the water as it is being filled to the brim in order to avoid the escape of dissolved radon from the samples. After collection, the samples were immediately taken to the laboratory [26]. In addition, since the samples would readily give up radon at

higher temperature, they were taken in an ice-containing bag to ensure that a low temperature was maintained. This is done to achieve maximum accuracy and not to allow the composition of the samples to change.

Radon concentration determination

As shown in Fig. 2, the water samples obtained from the mining site were analysed using a well-calibrated DurrIDGE RAD7-Active Electronic detector big bottle system (in this study, a 1.5 l plastic bottle was employed). The detector employs the method of alpha spectrometry in sample analysis. The Big Bottle System was adopted as it has the capacity to analyse large water samples of up to 2.5 l. The Big Bottle System employs a closed loop aeration process similar to that of RAD H₂O. In the closed loop aeration, the air volume and water volume are constant and independent of the flow rate. The air recirculates through the water and extracts the radon until a state of equilibrium develops. For the Big Bottle System, aeration period of more than 10 min is required to reach equilibrium as against the 5 min required in RAD H₂O system [25]. Before performing a measurement, the RAD7 was purged to be free of trapped radon and dry air. This was achieved through the laboratory drying unit. After purging the system, each 1.5 l of the collected water sample was coupled to the detector and the drying unit. The RAD7 was allowed to operate in Sniff mode to measure the radon concentration in the air loop and a conversion factor to radon in the water was applied [25]. The conversion factor employed was based on the configuration of

Fig. 2 A typical CAPTURE Big Bottle System configuration interface



the Big Bottle System (1.5 l plastic bottle) and temperature of the water at the time of the aeration. This was handled with the Durrigge's CAPTURE package as it makes it easy to obtain radon data and automatically calculate radon in water concentrations in the Big Bottle System [25].

Estimation of radon annual effective dose (RAED)

Radiation Dose from radon in drinking water can be received via ingestion or inhalation [27]. The ingestion and inhalation of radon in water poses a dangerous health risk by way of irradiation of radiosensitive cells in the human body once it is absorbed into the bloodstreams. Consequently, radon in drinking water could potentially cause other severe radiological health effects besides the lungs cancer that it is famous for, thus provoking human attention [28]. The radon annual effective dose ($\mu\text{Sv/y}$) for ingestion (RAED_{ing}) is estimated using Eq. 2 [1, 27].

$$\text{RAED}_{\text{ing}} = \text{CRn}_w \times C_w \times \text{EDC} \quad (2)$$

where RAED_{ing} implies the radon annual effective dose from ingestion ($\mu\text{Sv/y}$); CRn_w is the radon concentration in drinking water, C_w (equals to 0.6, 0.8, and 1.3 l/d for infant, children and adult respectively), is the annual water intake and EDC is the effective dose coefficient for ingestion and is equal to 7×10^{-8} , 2×10^{-8} , and 1×10^{-8} Sv/Bq for infants, children and adults respectively [27, 29]. It should be noted that for a given activity concentration of radon, the estimated dose can vary significantly depending on the water consumption rates and the dose conversion factors used. The annual consumption of water by individuals can vary significantly. A more conservative value of 1.3, 0.8 and 0.6 L per day for adults, children and infants respectively was adopted in this study as the water ingestion rates for the residents in the study area considering their climate, physical activity and culture.

To calculate the radon annual effective dose ($\mu\text{Sv/year}$) due to inhalation (AED_{inh}), Eq. 3 was applied [1, 27].

$$\text{RAED}_{\text{inh}} = \text{CRn}_w \times \text{Rn}_w \times F \times O \times \text{DCF} \quad (3)$$

where RAED_{inh} implies the annual effective dose from inhalation ($\mu\text{Sv/y}$); Rn_w is the ratio of radon in air to the radon in

water (10^{-4}); F is the equilibrium factor between radon and its progeny (0.4), O is the average indoor occupancy time per individual (7000 h/y) and DCF is the dose conversion factor for radon exposure (9 nSv/h (Bq/m^3)).

The total radon annual effective dose ($\text{RAED}_{\text{total}}$) derived from both ingested and inhaled dose summation as a result of utilization of the surface and ground waters in this mining area is calculated using Eq. 4

$$\text{RAED}_{\text{total}} = \text{RAED}_{\text{ing}} + \text{RAED}_{\text{inh}} \quad (4)$$

$\text{RAED}_{\text{total}}$ is the total radon annual effective dose ($\mu\text{Sv/y}$), RAED_{ing} implies the radon annual effective dose from ingestion ($\mu\text{Sv/y}$) and RAED_{inh} implies the radon annual effective dose from inhalation ($\mu\text{Sv/y}$).

Results and discussion

Table 1 presents the statistical summaries of the in-situ activity concentration of ^{238}U , Gamma radiation dose and the estimated annual effective dose. The complete results are available in "Data in Brief" [19]. Table 2 gives the correlational relationships between ^{238}U , DR and ^{222}Rn . The results of the Radon activity concentration and the corresponding effective dose absorbed from the surface and ground water samples are shown in Tables 3 and 4 respectively. Table 5 shows a comparative table of this present work and other literature reviewed.

Table 2 Pearson's correlation matrix showing the relationship between the measured ^{238}U , ^{222}Rn and the DR at the gold and lead mine field in Moro LGA

	DR (nGy/h)	^{238}U (Bq/kg)	^{222}Rn (Bq/kg)
DR (nGy/h)	1.0000		
^{238}U (Bq/kg)	0.2302	1.0000	
^{222}Rn (Bq/kg)	0.4230	0.8160	1.0000

Table 1 Measured activity concentration of ^{238}U , the DR and the annual effective dose (AED) from gold and lead mining area of Moro, Kwara State, North-Central Nigeria

	Min	Max	Mean	Median	SD	CV	Skew	Kurt	RL [1]
DR (nGy h^{-1})	56.30	90.30	77.11	77.05	7.87	10.21	-0.36	0.11	59.00
^{238}U (Bq kg^{-1})	3.71	97.57	43.47	44.46	23.89	54.97	0.16	-0.74	32.00
AED (mSv y^{-1})	0.07	0.11	0.09	0.09	0.01	10.86	-0.21	0.09	0.07

RL recommended limit, *min* minimum, *max* maximum, *SD* standard deviation, *CV* coefficient of variation

Table 3 ^{222}Rn concentration and annual effective dose in surface water from gold and lead mining area of Moro, Kwara State, North-Central Nigeria

STAT	^{222}Rn (Bq/l)	RAED _{ing} (μSv/year)			RAED _{inh} (μSv/y)	Total RAED (μSv/y)		
		Adult	Children	Infant		Adult	Children	Infant
MIN	10.28	48.78	60.04	157.59	25.91	74.68	85.94	186.87
MAX	21.77	103.30	127.14	333.73	54.86	158.16	182.00	379.67
MEAN	15.97	75.77	93.25	240.93	40.24	116.00	133.49	281.17
Median	15.43	73.22	90.11	236.54	38.88	112.10	128.99	271.77
Kurt	−1.74	−1.74	−1.74	−1.46	−1.74	−1.74	−1.74	−0.68
SKEW	0.04	0.04	0.04	0.25	0.04	0.04	0.04	0.24
SD	4.22	20.03	24.66	63.43	10.64	30.68	35.30	61.57
CV	26.44	26.44	26.44	26.33	26.44	26.44	26.44	21.90
RL [1]	11.10	100.00	200.00	—	—	100.00	200.00	—

RL recommended limit, *min* minimum, *max* maximum, *SD* standard deviation, *CV* coefficient of variation

Table 4 ^{222}Rn concentration and annual effective dose in ground water from gold and lead mining area of Moro, Kwara State, North-Central Nigeria

STAT	^{222}Rn (Bq/l)	RAED _{ing} (μSv/year)			RAED _{inh} (μSv/y)	Total RAED (μSv/y)		
		Adult	Children	Infant		Adult	Children	Infant
MIN	16.75	79.48	97.82	256.78	42.21	121.69	140.03	300.20
MAX	19.18	91.01	112.01	294.03	48.33	139.34	160.34	336.24
MEAN	17.98	85.32	105.01	275.82	45.31	130.63	150.32	321.13
Median	17.88	84.84	104.42	275.65	45.06	129.90	149.48	319.06
Kurt	−1.57	−1.57	−1.58	−1.56	−1.58	−1.57	−1.57	−1.14
SKEW	−0.04	−0.04	−0.04	−0.09	−0.04	−0.04	−0.04	−0.32
SD	0.87	4.11	5.05	13.25	2.18	6.28	7.23	12.64
CV	4.81	4.81	4.81	4.80	4.81	4.81	4.81	3.94
RL [1]	11.10	100.00	200.00	—	—	100.00	200.00	—

RL recommended limit, *min* minimum, *max* maximum, *SD* standard deviation, *CV* coefficient of variation

Results of the in-situ activity concentration of ^{238}U , DR and the estimated annual effective dose

The statistical summary of the measured activity concentrations of ^{238}U , DR and the annual effective dose for the 36 sample locations from gold and lead mining area of Moro, Kwara State, North-Central Nigeria is presented in Table 1. The measured values for all the parameters were moderately skewed because the distribution is moderately symmetric i.e. the measure of the asymmetry of their probability distribution about their means ranges between −1 and +1 [3, 20, 21, 30]. The activity concentrations of ^{238}U ranges between 3.71 and 97.57 Bq/kg with an average value of 44.46 Bq/kg. Surprisingly, this mean activity concentrations ^{238}U is higher than the global average of 32.00 Bq/kg provided by UNSCEAR [1]. This is a cause for worry as significant enhancement in the concentration of ^{238}U will increase the level of the background radiation and cause elevated amount of radon in the environment. Recall that ^{238}U available in the soil slowly disintegrates to other compounds such as radium, which over time eventually becomes radon. The values for the measured outdoor dose rate (DR) ranges between 56.30 and 90.30 nGy/h with an average value of 77.11 nGy/h. As

expected, this mean value for the DR is higher than the 59 nGy/h provided by UNSCEAR [1]. A correlation analysis was conducted to examine the relationship between the measured radionuclides and the DR. The result of the correlation analysis is given in Table 2. The results were classified as follows: $0.8 \leq |r| \leq 1$ denotes a strong correlation; $0.5 \leq |r| \leq 0.8$ denotes a significant correlation; $0.3 \leq |r| \leq 0.5$ denotes a weak correlation; and $|r| < 0.3$ denotes an insignificant correlation [3, 10, 20, 21]. A positive but non-significant correlation (0.2302) was observed between the ^{238}U and the DR. The positive correlation result suggests that the gold and lead mine field in Moro LGA is endowed with ^{238}U and it is contributing to the gamma doses received from the field.

The measured values of the DR at about 1 m above the ground level were used to evaluate the annual effective dose (AED) using Eq. 1. The resulting highest, lowest and mean values for the annual effective dose are 0.11, 0.07 and 0.09 mSv/y respectively. The estimated mean value is higher than the recommended limit of 0.07 mSv/y provided by UNSCEAR [1]. This follows that the risk of gamma radiation exposure at the gold and lead mine field in Moro LGA is high and the general public are not safe from overexposure to ionizing radiation.

Table 5 Comparison of radon concentration data obtained from other regions

Water sources	Radon activity concentration (Bq/L)	Region of study	References
Groundwater	0.168–78.509	Ijero, Nigeria	[18]
Drinking water	3.176–49.932	Shanono and Bagwai, Nigeria	[9]
Groundwater	7.41 ± 2.04	Zaria, Nigeria	[33]
Groundwater	7.002 ± 0.33	Kaduna, Nigeria	[34]
Spring and non-bottled mineral waters	1.4–105	Spain	[35]
Groundwater	3043	Poland	[36]
Groundwater	17–3856	Portugal	[37]
Tap water	0.46 ± 0.04	Iran	[38]
Ground water	2.1–206.1	Ekiti, Nigeria	[39]
Surface water	≤ 10	Worldwide	[32]
Ground water	≤ 10	Worldwide	[32]
Surface water	≤ 11.1	Worldwide	[31]
Ground water	≤ 11.1	Worldwide	[31]
Surface water	10.28–21.77	Kwara, Nigeria	This study
Ground water	16.75–19.18	Kwara, Nigeria	This study

Radon concentration in surface and ground water samples

The measured ^{222}Rn activity concentration varied from 10.28 to 21.77 Bq/l with a mean value of 15.97 Bq/l for surface (stream) water and 16.75 to 19.18 Bq/l with a mean value of 17.98 Bq/l for ground water, respectively. Observations from the surface samples shows that most (64%) of the results are higher than the limit of 11.1 Bq/l [31]. Expectedly, the overall mean values for the surface water samples which was found to be 15.97 Bq/l is greater than the maximum contaminant level of 10 and ~ 11.1 Bq/l given by both WHO [32] and US-EPA [31], respectively.

Ground water samples, on the other hand, have 100% of its values above the limits set by both US-EPA and WHO with an overall arithmetic mean of 17.98 Bq/l. Ground water samples have obviously higher values as compared to surface samples. This could be due to the fact that radon has higher chance to escape the surface than to accumulate while it has higher chances to accumulate in ground water due to contact with geologic formations which is rich in uranium [15]. Considering the higher values of ^{238}U measured using Super Spec RS-125 gamma spectrometer, it was not surprising to have higher radon activity concentration. In fact, a very strong positive correlation (0.8160) was observed between the ^{238}U and the ^{222}Rn (see Table 2). In the same vein, higher concentrations in the surface and ground water

samples could be due to the low partition coefficient of radon in surface water which reduces the rate of degassing. Surface samples show 26.44% coefficient of variation while ground samples had 4.81% coefficient of variation. This shows a very low dispersion rate in the radon activity concentration obtained from ground water compared to surface water.

Table 5 shows the variation in the values obtained for radon activity concentration as compared with ones obtained at different regions and countries. The concentrations were higher than some of the literatures reviewed and generally exceeded the set limits of both US-EPA and WHO. This could pose a threat especially serious cancer risk on the residents in the immediate environment due to possible risk of exposure to high radon concentration either by ingestion or inhalation. Hence, a need to calculate the annual effective dosage of radon is desired.

Radon annual effective dose in surface and ground waters

The values of the annual effective doses calculated are also presented in Tables 3 and 4. This is used to ascertain the possible risk incurred due to exposure to this radon gas. This is necessary because a careful study of the local resident's life-style reveals that they drink directly from the water sources without any form of purification process. The values of the radon annual effective dose due to ingestion ranges from 48.78 to 103.30 with a mean of 75.77 $\mu\text{Sv/y}$, 60.04 to 127.14 with a mean 93.25 $\mu\text{Sv/y}$ and 157.59 to 333.73 with a mean of 240.93 $\mu\text{Sv/y}$ for adult, children and infants, respectively for surface water. And 79.48 to 91.01 $\mu\text{Sv/y}$ with a mean of 85.32 $\mu\text{Sv/y}$, 97.82 to 112.01 $\mu\text{Sv/y}$ with a mean of 105.01 $\mu\text{Sv/y}$ and 256.78 to 294.03 $\mu\text{Sv/y}$ with a mean of 275.82 $\mu\text{Sv/y}$ for adult, children and infants, respectively for ground water. In addition, the radon annual effective dose due to inhalation ranges from 25.91 to 54.86 $\mu\text{Sv/y}$ with a mean of 40.24 $\mu\text{Sv/y}$ and 42.21 to 48.33 $\mu\text{Sv/y}$ with a mean of 45.31 $\mu\text{Sv/y}$ for surface and ground water samples, respectively.

The total radon annual effective dose obtained from the summation of both RAED_{ing} and RAED_{inh} for surface waters ranges from 74.68 to 158.16 $\mu\text{Sv/y}$ with a mean of 116.01 $\mu\text{Sv/y}$, 85.94 to 182.00 $\mu\text{Sv/y}$ with a mean of 133.49 $\mu\text{Sv/y}$ and 186.87 to 379.67 $\mu\text{Sv/y}$ with a mean of 281.17 $\mu\text{Sv/y}$ for adult, children and infant respectively. Consequently, for ground water, the values ranges from 121.69 to 139.34 $\mu\text{Sv/y}$ with a mean of 130.63 $\mu\text{Sv/y}$, 140.03 to 160.34 $\mu\text{Sv/y}$ with a mean of 150.32 and 300.20 to 336.24 $\mu\text{Sv/y}$ with a mean of 321.14 $\mu\text{Sv/y}$ for adult, children and infant, respectively. Comparing the mean $\text{RAED}_{\text{total}}$ ($\mu\text{Sv/y}$) obtained from both waters with 100 $\mu\text{Sv/y}$ and 200 $\mu\text{Sv/y}$ limits for adults and children respectively given by WHO [30], it was discovered that the RAED ($\mu\text{Sv/y}$) for children was at the safe range, however, that of the adult poses serious radiation and cancer

risk as it is above the safe limit. The values obtained for infant was generally high compared to that of adult and children and this poses serious health risk as their developing critical organs are very radio-sensitive compared to that of adults and children that had developed better.

Conclusion

This work reported the activity concentration of ^{238}U , the DR, and the ^{222}Rn concentration levels in ground and surface water samples from gold and lead mining area of Moro, Kwara State, Nigeria. The results show that the risk of radiation exposure at the gold and lead minefield in Moro LGA is high and the general public are not safe from exposure to ionizing radiation. Although, the radon annual effective dose for children was found to be lesser than $200\text{ }\mu\text{Sv/y}$ recommended for children, proper monitoring on the mining activities is encouraged since no amount of radiation is safe for stochastic effects. It is therefore recommended that water in this environment should be treated properly before being utilised.

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Author contributions MMO conceived and designed the research work, collect the data and wrote the paper. FCA, TBA, ODA, OO and MOM performed the risks analysis and final compilation of the work.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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