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RESEARCH PAPER

Annual Effective Dose Assessment of Radon in Drinking Water from Abandoned Tin and Cassiterite Mining Site in Oyun, Kwara State, Nigeria

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ABSTRACT:

Mining activities are generally known to enhance the concentration of primordial radionuclides in the environment thereby contributing immensely to human exposure to ionizing radiation of terrestrial origin. Thus, the abandoned Tin and Cassiterite mining site in Oyun, Kwara State, Nigeria, is believed to cause radiological implications on local residents. Assessment of radon concentration in surface water from the study area was carried out using RAD7-Active Electronic detector big bottle system. In order to ascertain the risk or hazard incurable in consuming such water, 12 samples were analysed and used in the estimation of annual effective dose of radon. The measured maximum and minimum radon concentrations were found to be 44.95 and 21.03 Bq/L with average of 35.86 Bq/L. These values are quite greater than the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) recommended limits of 11.1.Bq/L. The estimated total effective dose ($AEDE_{total}$) was found to be within the range of 206.52 and 441.41 μSvy^{-1}, and an average of 352.20 μSvy^{-1} for Adults, 283.30 and 605.47 μSvy^{-1}, and average of 483.10 μSvy^{-1} for Children, and finally, 321.70 and 687.47 μSvy^{-1} with average of 548.64 μSvy^{-1} for Infants, respectively. These values were higher than the recommended limit of 100 μSvy^{-1} and 200 μSvy^{-1} for adult and children respectively. Furthermore, worries should be noted about the probabilistic cumulative effect on the consumers of such water if the ingestion is for an extended period of time.

keywords: Tin, Cassiterite, Mining, Radon, Annual Effective Dose.

INTRODUCTION

Geologically, Nigeria is hugely blessed with diverse natural resources (Salawu *et al*., 2020, 2021). Over the time, mankind effectively invented strategies to exploit these minerals for development. Mining is the deep excavation of the subsurface in order to extract useful mineral resources below, buried by overburden. The mining industry is seen as one of the major supporters of most economic growth and development processes in many nations with Nigeria not being an exception (Orosun, 2021; Bradshaw, 2005). Presently, mining in Nigeria is carried out partly by illiterate local artisans and the crude methods adopted by these artisans

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exposes them and the immediate environments to dust and other earth materials, containing high levels of radionuclides and heavy metals (Adagunodo *et al*., 2019; Usikalu *et al*., 2020; Bello *at al*., 2020; Orosun, 2021; Orosun *et al*., 2020, 2020a). The radionuclides brought to the surface through mining, find their way to different water bodies, air spaces, soils; and breaks down spontaneously to radium and eventually radon. Radon is a naturally occurring radioactive gas without taste, smell or color and it is chemically unreactive (ISO, 2013; ICRP, 2010). Radon and its progenies exist as isotopes which are produced through alpha decay of Radium, depending on the parent radionuclide (Ajibola *et al*., 2021). ^{238}U decays to give ^{222}Rn having a half-life of 3.82 days; while ^{235} U and ^{232}Th decays to give ^{219}Rn and ^{220}Rn with half-lives of 4 s and 55.6 s respectively. Radon is present almost everywhere on earth in traceable amounts and it is distributed in the soil, groundwater and in the lower region of the atmosphere (ISO, 2013; ICRP, 2010; USEPA, 2003; Oni *et al*., 2021). Studies have shown that factors such as geological state, geochemical properties of the parent radionuclide, hydrological state and the quantity or amount of the parent radionuclides are the likely parameters that are capable of influencing the concentration and distribution of Radon (Orosun *et al*., 2021; Bello *et al*., 2020; Usikalu *et al*., 2020; Adagunodo *et al*., 2019; Akinnagbe *et al*., 2018; Ravikumar and Somashekar, 2014; Manzoor *et al*., 2008).

Water is a universal solvent, so essential to mankind for consumption, domestic purposes and industrial purposes. Sources of water can be classified into two categories which are surface and groundwater. Surface waters include water from rivers, lakes, seas, as well as big ponds from abandoned mines. Radon is soluble in water and it may be found in appreciable quantities in surface water due to the presence of radium in the water, bedrock and surrounding soil (USEPA, 2003). The concentration of radon in surface water are generally very low but can be elevated by anthropogenic activities such as mining (Orosun *et al*., 2018, 2021; Ajibola *et al*., 2021). Relatively high concentrations of radon in water from bedrocks are found frequently in areas where the bedrock consist of granites and is surplus in Uranium (Akerblom and Mellander, 1997). When radon-contaminated water is used for indoor purposes, the dissolved radon in the water outgases and becomes airborne. Radon has a half-life of 3.82 days, during its disintegration it gives off alpha particles successively yielding, Polonium, Bismuth and Lead. This poses great harm to human health. Approximately 54% of the total internal radiation that humans are exposed to is contributed by Radon (ICRP, 1993). Radon is believed to be the second largest cause of lung cancer, following smoking (USEPA, 1999, 2003; UNSCEAR, 2000). This paper investigates the concentration of radon in water from the abandoned Tin and Cassiterite mining site in Oyun, Kwara State, for the assessment of the health risks on the consumers. The aim of the work is to estimate the annual effective dose via ingestion and inhalation and compare with regulatory standards while adding to the database of radon concentration of drinking water sources in Nigeria.

MATERIALS AND METHODS

Oyun is one of the 16 Local Government Area (LGA) in Kwara State, Nigeria. It is geographically positioned on longitude 4° 42' 0" East and latitude 8° 7' 0" North. Its headquarters is located situated at Ilemona. The landmass of Oyun span approximately 476km^2 with a population of about 94,253 as at 2006 census. The population was made up 47,890 males and 46,564 females with 40,835 within 0-14 years, 50,419 within 15-69 years and 3,200 are 70 years and above. As at 2016, the total population was projected to be around 127,500. It is positioned in the transition zone between the savannah and forest regions of the State. Its climatic condition is of the wet and dry tropical type having an average yearly rainfall of approximately 1,318 mm and an average monthly temperature of approximately

32°C. The maximum temperature is witnessed in the month of March. This LGA receives rainfall from the south-western air masses, which enters Nigeria from the Gulf of Guinea coast (Akinbile, *et al.*, 2006).

Geologically, the Nigerian Basement Complex of southwestern Nigeria surrounds Kwara State. The region is underlain by the Migmatite–Gneiss complex of Precambrian age, which is intruded by older granitic bodies also known as the Pan-African Granitoids. Younger metasedimentary rock units, probably Paleo-Protozoic age, underlie parts of the study area. Migmatite–Gneiss complex rock units constitute almost 75% of the rock units occurring within the region, which includes Migmatite Gneiss, Banded Gneiss, Granite Gneiss and Quartzite (Orosun *et al*. 2019, 2020a&b). The Younger Metasedimentary rocks constitute almost 25% of the remaining rocks of the study area (Balogun 2019). These Granite Gneiss and Quartzite from the study area have been reported to be rich in minerals and contains elevated amount of natural radionuclides (Orosun *et al*., 2019). Tin and Cassiterite mineralization is mostly found to be linked with gneisses and schists in this part of the country. This suggests that the area of mineralization is extensive and so are the mining activities.

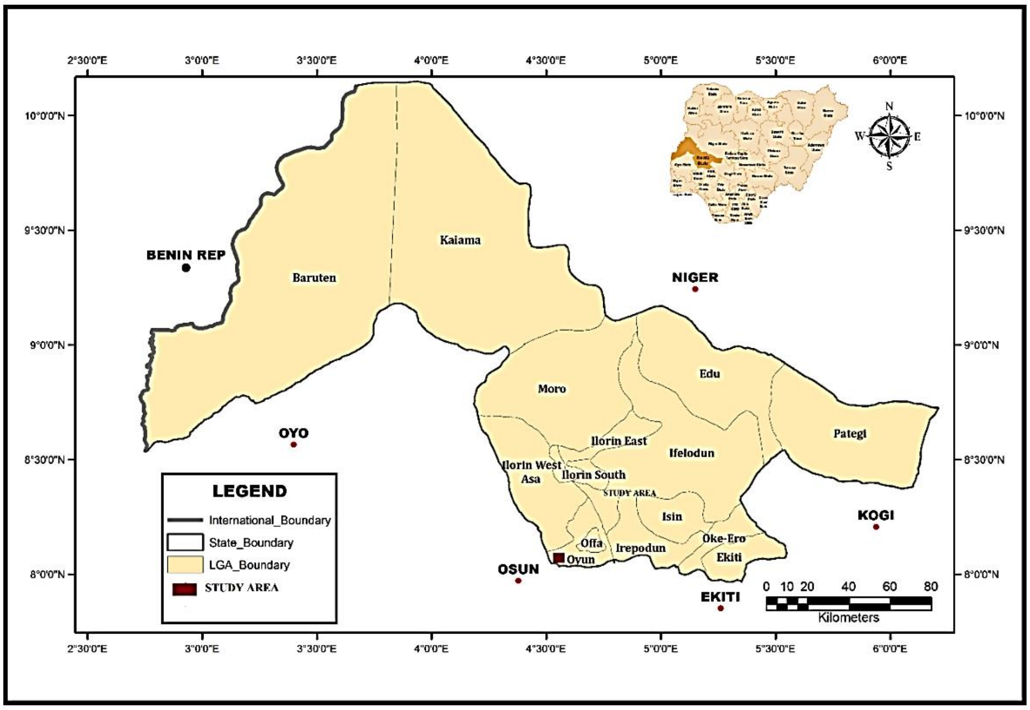
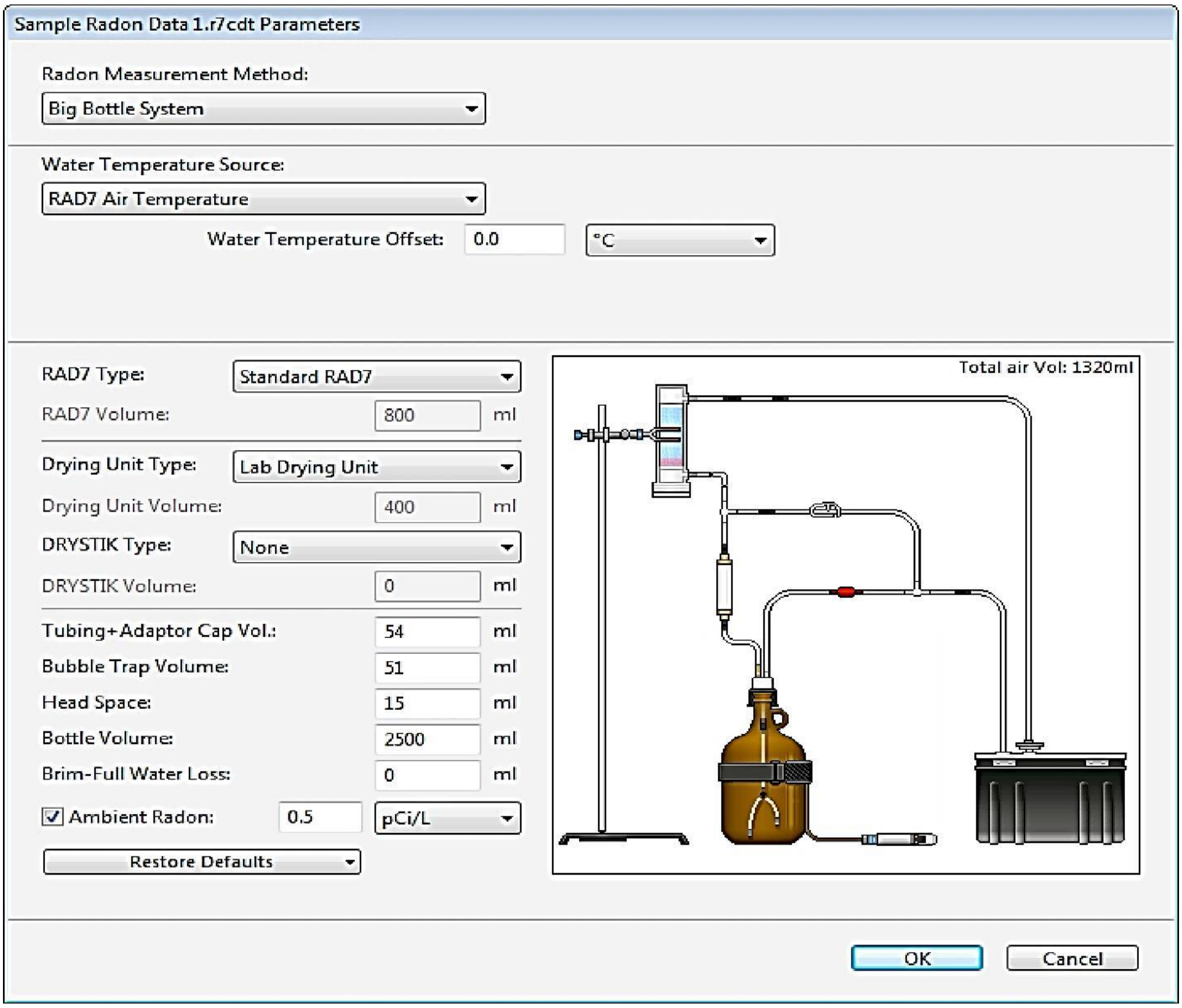


Fig 1: Show the geological map of Oyun in Kwara State which is the study area

12 surface water samples were collected from the abandoned Cassiterite and Tin mining site located in Oyun, Kwara State, Nigeria. These samples were collected early in the morning so that certain properties of the water will remain unchanged and not disturbed by human activities. It should be noted that the local inhabitants living in the vicinity of the mines depends on the untreated waters from the abandoned mine for drinking. The samples were collected in a 1.5 L bottle because the cork of the RAD7 water kit can fit well into the mouth of the bottle. Water samples were collected directly from the source into the clean 1.5 L bottle that has been previously rinsed with distilled water. During the process of water collection, the samples from the surface water were collected by immersing the bottle directly inside the fetcher and when it is filled to the brim, the bottle is covered inside the water while still in the fetcher. This is done in a bid to minimise the escape of dissolved radon in the water (Ajibola

et al., 2021). The samples were then taken to the laboratory immediately after collection for analysis to ensure maximum precision and to avoid variation in the composition of the sample (Isinkaye and Ajiboye, 2017).

A well calibrated Durridge RAD7-Active Electronic detector big bottle system (as indicated in Figure 3, a 1.5 l plastic bottle was employed) was used to analysed twelve (12) samples of the surface waters collected from the abandoned Tin and Cassiterite mine pit in Oyun, Kwara State, Nigeria. The Durridge RAD7 big bottle system employs a closed loop aeration method that were similarly employed in the commonly used RAD H_2O (Orosun *et al.*, 2021; Ajibola *et al.*, 2021). In this method (closed loop aeration), the volumes of water and air are kept at stable levels and independent of the flow rate. The air flows repeatedly through the water body extracting the radon until equilibrium state is established. This electronic detector make use of alpha spectrometry method. It can accurately give the radon concentration levels in water samples in just about 20 min. In comparison, this measurement period is appreciably lees than 3.8 days, which is the half-life of radon. This consequently makes the Durridge RAD7-Active Electronic detector superior to several detectors for the measurement of radon in water.



Sample Radon Data 1.r7cdt Parameters

Radon Measurement Method: Big Bottle System

Water Temperature Source: RAD7 Air Temperature

Water Temperature Offset: 0.0 °C

RAD7 Type: Standard RAD7

RAD7 Volume: 800 ml

Drying Unit Type: Lab Drying Unit

Drying Unit Volume: 400 ml

DRYSTIK Type: None

DRYSTIK Volume: 0 ml

Tubing+Adaptor Cap Vol.: 54 ml

Bubble Trap Volume: 51 ml

Head Space: 15 ml

Bottle Volume: 2500 ml

Brim-Full Water Loss: 0 ml

☑ Ambient Radon: 0.5 pCi/L

Fig 3: A typical CAPTURE Big Bottle System configuration interface

In this work, the water samples were taken to the laboratory immediately after collection in order to avoid the decay coefficient; hence, no decay-correction was applied (Isinkaye &

Ajiboye, 2017). The factor of conversion utilized was established on the ground of the configuration of the RAD7-Active electronic big bottle system (1.5 l plastic bottle) and temperature of the sampled drinking water at the time of the aeration. This was ensured using the Durridge's CAPTURE package that acquires radon data and automatically calculate radon in water concentrations in the RAD7-Active electronic big bottle system. More detail on the procedures can be found in our earlier works (Orosun *et al.,* 2021a; Ajibola *et al.,* 2021).

RESULTS AND DISCUSSION

Table 1 and Figure 4 below shows the Radon Concentrations in the water samples obtained from the mining site. The statistical summary of the results of the measured activity concentrations of ^{222}Rn for the polluted mine pool is presented in Table1 as well. Descriptive statistics consisting of the lowest value (Min.) and highest value (Max.), Skewness, arithmetic mean, standard deviation (SD) and the coefficient of variation (CV) were explored using IBM SPSS version 25 package operated on Window 10 operating system. The results revealed that the radon activities in the surface water samples were normally distributed since the measure of the asymmetry of the probability distribution about the means is -0.7949 (comfortably within the range of -1 and +1) (Normality Testing, 2019; Orosun *et al*., 2020b). The coefficient of variation (CV) evaluation shows low variability in the distribution of radon concentration in the surface water. Since $CV \leq 20\%$ implies low variability, $20 < CV \leq 50\%$ indicates medium, and $50\% < CV \leq 100\%$ represents high variability.

The estimated mean concentration of radon in surface water was observed to range from 21.03 Bq/L to 42.19 Bq/L. The highest radon concentration of 42.19 Bq/L was recorded in sample OYSW11 and the least value of 21.03 Bq/L was recorded in sample OYSW12. All the samples analysed had radon concentrations above the maximum permissible limit of 11.1 Bq/L (USEPA, 2003) and the world average value of 10 Bq/L (WHO, 2011). However, none of the measurements was up to 100 Bq/L which is the maximum contaminant level (MCL) recommended by European Union for measurement that warrants consideration of possible remedial actions and 1000 Bq/L recommended European Union upper bound value above which remedial action is definitely required (Aruwa *et. al.,* 2017).

Table 1: ^{222}Rn concentration in surface water from Tin and Cassiterite mining site in Oyun, Kwara State, Nigeria.

Sample Code	**Source**	**Rn-222 (Bq/L)**	**Error ±**
OYSW1	Surface	40.31	7.12
OYSW2	Surface	39.72	5.94
OYSW3	Surface	43.88	6.12
OYSW4	Surface	41.91	7.09
OYSW5	Surface	33.84	4.87
OYSW6	Surface	31.72	3.95
OYSW7	Surface	44.95	8.87
OYSW8	Surface	40.08	7.23
OYSW9	Surface	27.93	4.58
OYSW10	Surface	22.83	4.01
OYSW11	Surface	42.19	5.89
OYSW12	Surface	21.03	3.11
Min		21.03	3.11
Max		44.95	8.87
Skewness		-0.7949	0.196312
Mean		35.8658	5.731667
SD		8.25692	1.683243
CV		4.34373	3.405133

The overall estimated mean radon concentration in the drinking water was 35.87Bq/L, which was also observed to be greater than the maximum permissible limit of 10 Bq/L and 11.1 Bq/L given by WHO and USEPA (WHO, 2011; USEPA, 2003) respectively. These high values were attributed to the bedrock's geological structure of the study area that was explored by mining activities. Recent researches has shown that ^{222}Rn is the principal cause of lung cancer among non-smokers of tobacco, thereby constituting deleterious risk to humankind (Ajibola *et al*., 2021; Bello *et al*., 2020)

Comparatively, even though the values reported in this current study are higher than the values recommended by WHO and USEPA, they agree with the findings of Ajibola *et al*. (2021) and Orosun *et al*. (2021) for mining locations in other parts of the Kwara State, Nigeria.

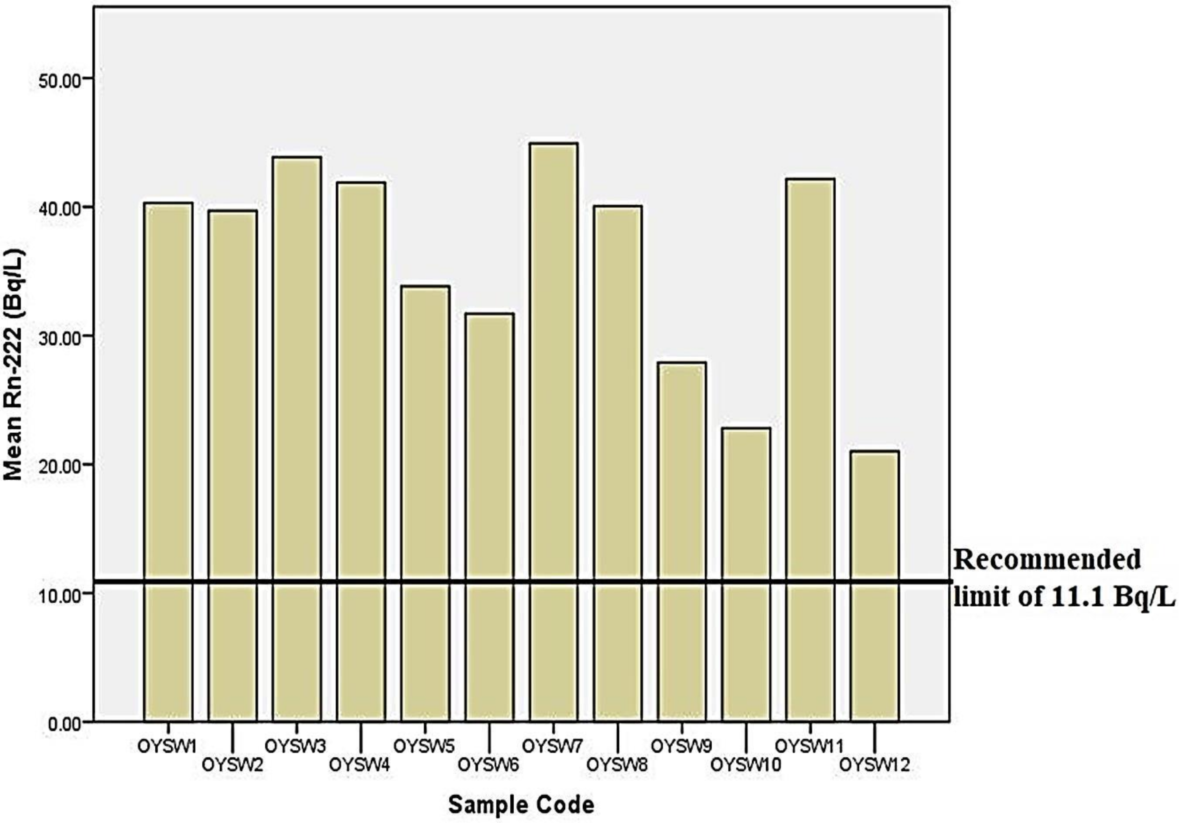


Fig 4: Radon Concentrations in the surface water samples obtained from the mining site.

This radon present in drinking waters gets into the body through ingestion or inhalation (during degassing). During inhalation or ingestion, it degenerates to its progenies giving out radiation such as alpha particles that irradiates the lungs or the stomach’s cells, inducing cancer.Radiation dose estimated from the presence of radon in drinking water was calculated via the ingestion pathway. However, radon gas present in the drinking water can also find their way into the indoor air in the course of bathing and other domestic usages causing substantial rise in the risk of lung cancer because of inhalation of radon.

The annual effective dose (µSv/y) due to ingestion of the drinking water was calculated using:

$$AEDE_{ing} = K \times C_{Rn} \times C_w \times T \quad (1)$$

where K is the conversion factor for ingesting dose of ^{222}Rn (Sv Bq$^{−1}$). For adults, committed effective dose per unit intake from the ingestion of radon in water (K) is $10^{−8}$ Sv Bq$^{−1}$. It is 2 × $10^{−8}$ Sv Bq$^{−1}$ and 7 × 10−8 Sv Bq$^{−1}$ for children and infants respectively. C_w is the water consumption. Estimated as 2L per day for adults, 1.5L per day for children and 0.5L per day for infants. C_{Rn} is the concentration of ^{222}Rn (BqL$^{−1}$) in each sample and T is the time span of consumption (365 days) y^{-1}.

To calculate the $AEDE_{ing}$, we assumed that adult, children and infant drinks directly from the source and consumes an average of 2.0, 1.5, and 0.5 L of water per day, respectively (WHO, 211; Duggal, *et al.*,2013; Bem *et al.*, 2014).

To calculate the AEDE via inhalation we used equation 2 giving as:

$$AEDE_{inh} = C_{Rn} \times F \times O \times DCF \qquad (2)$$

where $AEDE_{inh}$ is the Annual effective dose via inhalation (μSv/y); R_{nw} 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 (7,000 h. y^{-1}). DCF is the dose conversion factor for Radon exposure (9 nSvh^{-1}(Bqm-3)$^{-1}$).

The total Annual Effective Dose ($AEDE_{total}$) 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 equation 3

$$\mathrm{AEDE_{total}} = \mathrm{AEDE_{ing}} + \mathrm{AEDE_{inh}} \qquad (3)$$

AED_{total} is the total annual effective dose (μSvy^{-1}), AED_{ing} implies the Annual effective dose from ingestion (μSvy^{-1}) and AED_{inh} implies the Annual effective dose from inhalation (μSvy^{-1}).

Table 2 and Figures 5 and 6 gives the estimated annual effective doses (μSv/year) via ingestion and inhalation respectively as well as the total annual effective doses for adults, children and infants. The average AEDE (μSv/year) for the ingestion of water by adults, children and infants were estimated to be 261.82, 392.72 and 458.26 μSv/year respectively. The AEDE for inhalation span from 52.99 to 113.27μSv/year with an average of 90.39 μSv/year. The average of the Total AEDE (for both ingestion and inhalation) for the water sample were estimated to be 352.20, 483.10 and 548.64 μSv/year for adults, children and infants respectively. These values are higher than the WHO permissible limit of 100 μSv/year and 200 μSv/year for adults and children respectively (WHO, 2011) thereby revealing high level of radiological health risks for all age groups. From Figures 5 and 6, it is evident that the order of radiation dose to all age groups follows the order infant > children > adult. Thus, the values obtained for infant was higher than the adult and children values. This constitutes deleterious health risk to them because their critical organs are not fully developed and are therefore very sensitive to radiation compared to that of adults and children that had developed better (Orosun *et al.*, 2021).

Table 2: The Annual effective dose estimate for ingestion pathway (µSv/year).

Sample Code	Source	$AEDE_{ing}$ (µSv/year)			$AEDE_{inh}$ (µSv/year)	$AEDE_{total}$ (µSv/year)		
		Adults	Children	Infants		Adult	Children	Infants
OYSW1	Surface	294.26	441.40	514.96	101.58	395.84	542.98	616.54
OYSW2	Surface	289.96	434.90	507.40	100.09	390.05	534.99	607.49
OYSW3	Surface	320.32	480.50	560.60	110.58	430.90	591.08	671.18
OYSW4	Surface	305.94	458.90	535.40	105.61	411.55	564.51	641.01
OYSW5	Surface	247.03	370.50	432.30	85.28	332.31	455.78	517.58
OYSW6	Surface	231.56	347.30	405.20	79.93	311.49	427.23	485.13
OYSW7	Surface	328.14	492.20	574.20	113.27	441.41	605.47	687.47
OYSW8	Surface	292.58	438.90	512.00	101.00	393.58	539.90	613.00
OYSW9	Surface	203.89	305.80	356.80	70.38	274.27	376.18	427.18
OYSW10	Surface	166.66	250.00	291.70	57.53	224.19	307.53	349.23
OYSW11	Surface	307.99	461.98	539.80	106.32	414.31	568.30	646.12
OYSW12	Surface	153.52	230.30	268.70	52.99	206.52	283.30	321.70
MIN		153.52	230.30	268.70	52.99	206.52	283.30	321.70
MAX		328.14	492.20	574.20	113.27	441.41	605.47	687.47
SKEW		-0.795	-0.7944	-0.7941	-0.795	-0.795	-0.794	-0.794
MEAN		261.82	392.72	458.26	90.38	352.20	483.10	548.64
SD		60.28	90.41	105.53	20.81	81.08	111.22	126.33

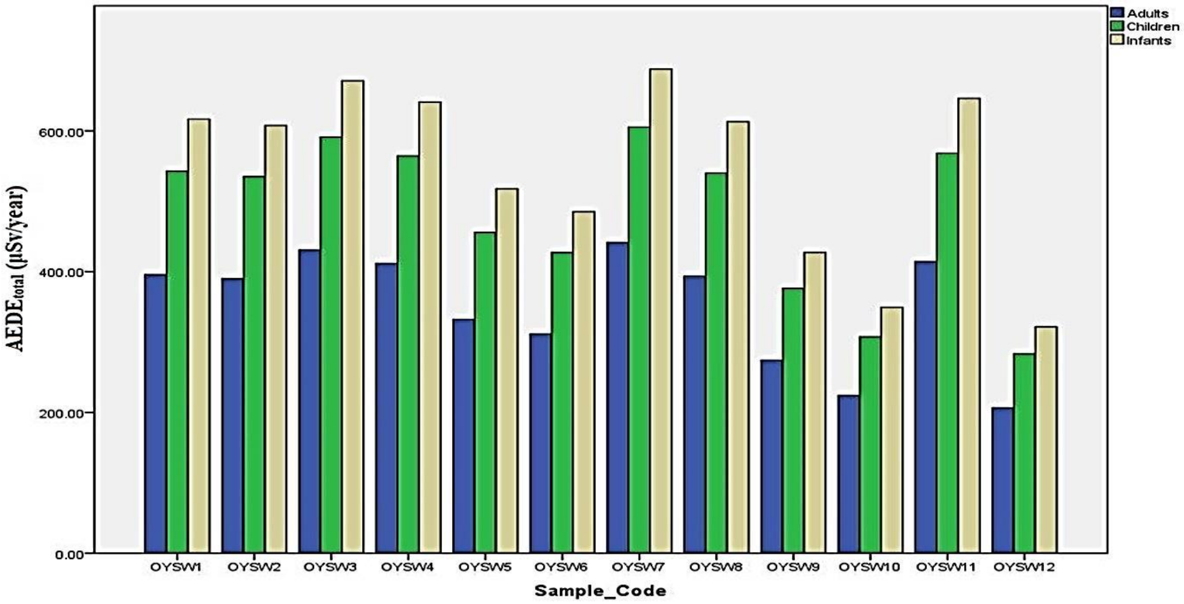


Fig 5: The estimated $AEDE_{total}$ (µSv/year) for the different age group for all the sampling locations.

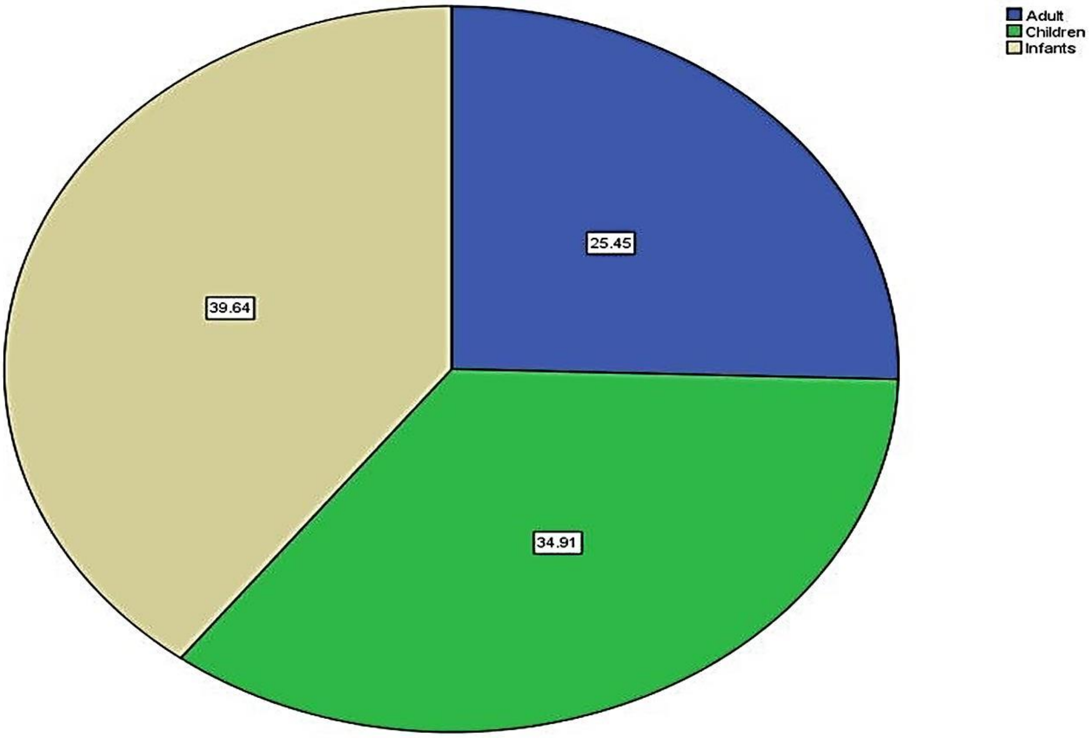


Fig 6: Estimated mean $AEDE_{total}$ (μSv/year) for the different age group.

CONCLUSION

A study of Radon concentration in surface water has been carried out at abandoned Tin and Cassiterite mining site in Oyun, Kwara State, Nigeria, using a calibrated RAD7-Active electronic big bottle system. This was done in order to ascertain the radiological safety of using the water for drinking and other domestic purposes. The mean Radon concentration and the corresponding annual effective doses for adults, children and infants showed that the water from this mine pit is not radiologically safe as the values were mostly above the recommended limits set by regulatory bodies. The order of estimated annual effective dose for both surface and ground waters follow the pattern infant > children > adult.

It is known that in Nigeria until date no known standard regulations of radon concentration in water have been developed. Therefore, there is a need to develop and establish national maximum contaminant limit of radon and other radionuclides in drinking water. In order to achieve this, radon concentration in all drinking water sources should be investigated across all geo-political zones in Nigeria. This will help to investigate radon risk areas and seek for ways to protect these citizens from risks associated with the inhalation and ingestion of radon.

In addition, public water system should be revisited and efforts should be made to educate and enlighten the public on radon, its health effects and remedial actions necessary to reduce radon concentration in water. Inhalation and ingestion of radon have been associated with incidence of stomach and lung cancer. Hence, there is need to carry out epidemiological studies to investigate the incidence of lung and stomach cancer in the study area and other areas in Nigeria where high radon concentration is observed.

GRANT SUPPORT DETAILS

The present research did not receive any financial support.

CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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